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EFFECT OF PROTON CHANNELING AT 2.8 MeV ON THE $Cu^{65}(p,n)Zn^{65}$ REACTION RATE IN A SINGLE CRYSTAL OF Cu

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When a beam of ions is incident on a crystal, penetration is enhanced if the direction of incidence is parallel to either a low-index direction or a low-index plane. For heavy ions in the 1- to 100-keV range of energies, this was demonstrated by the experiments of Davies et al.[1] and Nelson and Thompson.[2] The latter experiments extended to the light ions H^+ and He^+. Similar experiments with protons, deuterons, and alphas in the MeV range by Dearnaley,[3] Dearnaley and Sattler,[4] and Erginsoy, Wegner, and Gibson[5] show that the energy loss of some particles passing through a thin Si crystal is anomalously low when incidence is close to low-index planes and directions.

Near these critical orientations the ion's trajectory must find the atoms aligned in rows or sheets, depending on whether incidence is parallel to a direction or a plane. One interpretation of the effect is that a succession of glancing collisions steers the trajectory so that it becomes channeled between the rows or sheets.

The keV heavy-ion case was predicted by Robinson and Oen[6] from machine calculations of recoil ranges in irradiated solids. Analytical treatments by Lehmann and Leibfried[7] and myself[2] confirm that for reasonable ion-atom potentials the effect should exist. The latter calculations can easily be extended to higher energies and show that channeling should still occur at MeV energies.

If the channeling hypothesis is correct then the ion passes through a region of the crystal where its chance of meeting a nucleus is greatly reduced. One should therefore expect a decrease in nuclear reaction rate when a beam of initiating charged particles is incident along a channel direction. Such an experiment has been performed at Harwell and is described here.

At a recent Symposium, this experiment was reported simultaneously with one by Davies and the Aarhus group[8] who had independently found an effect with a (p,γ) reaction in Si at 410 keV.

The reaction chosen for the experiment was $Cu^{65}(p,n)Zn^{65}$, which has a threshold near 2.1 MeV and a cross section rising through 0.1 barn at about 3 MeV. In making this choice it was hoped that a significant fraction of protons entering a suitably oriented Cu crystal would become channeled, and would slow down below 2.1 MeV with a reduced chance of reactive nuclear encounters. A Cu crystal was therefore bombarded with 2.8-MeV protons and the rate of neutron emission studied as a function of orientation.

The crystal was roughly 1 cm thick so that all protons were stopped in the target. An ion following a normal trajectory would travel about 10^{-3} cm in falling from 2.8 to 2.1 MeV. The surface normal was about 5° from the $\langle 110 \rangle$ direction, and the crystal was clamped into a holder that could be rotated about this surface normal through 360° with a precision of better than 0.1°. This axis was horizontal, and rotation about it will be denoted by the angle θ. The proton beam was also horizontal, and the angle φ between it and the θ axis (surface normal) could also be varied to a precision of better than 0.1°. The shape of the crystal holder restricted the range of φ to ±70°. This geometry is illustrated in Fig. 1.

Consider the effect of a change $\Delta\theta$ in θ for a fixed φ. The direction in the crystal OP, which was initially along the proton beam, moves through an angle $\Delta\Psi$ to the position OQ. It is easily shown that

$$\Delta\Psi = \sin\varphi \Delta\theta.$$

Thus when φ is small, the change in orientation $\Delta\Psi$ for a given $\Delta\theta$ can be extremely small and this allows angular effects to be investigated to a very fine precision. For example, with $\varphi = 5°$ and $\Delta\theta = 0.1°$, $\Delta\Psi = 0.01°$.

The orientation of the crystal is best described with reference to the stereogram in Fig. 2 which has the θ axis as pole and shows a limited region out to $\varphi = 10°$ that includes the $\langle 110\rangle$ direction. A particular direction of incidence is represented by a point on the stereogram, and if θ is varied at constant φ, the locus of this point is a circle centered on the pole $\varphi = 0$. In the experiment two runs were made with constant φ at 5.0° and 5.4°, and a third run was made with θ constant at 255°. The regions scanned are indicated by dotted lines on the stereogram. One should note that the initial orientation of the crystal was obtained from a Laue diffraction pattern using a standard x-ray camera. The uncertainty in initial orientation was therefore quite large (±0.5°) compared with the precision to which changes in orientation could be made.

The proton beam came from the Harwell 5-MeV Van de Graaff accelerator. After passing through a 90° analyzing magnet it was collimated by a tantalum aperture to have an angular divergence not greater than ±0.075°. In order to find the position $\varphi = 0$, when the θ axis lies along the beam, the crystal was replaced by a flat plate of glass and optical reflections from this were used to align the normal with

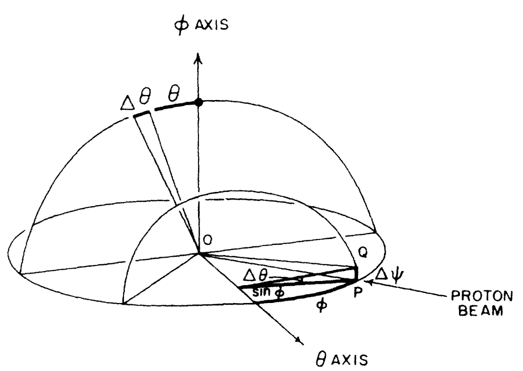

FIG. 1. The geometry of the crystal and proton beam.

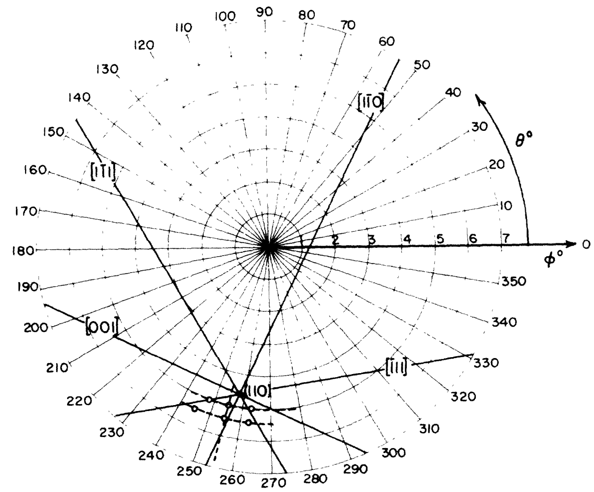

FIG. 2. Stereogram, with the incidence axes as pole, showing by dotted lines how the crystal orientation was changed in the three runs of Fig. 3.

the beam collimator.

Neutrons were detected by two BF_3 proportional counters, each embedded in a separate paraffin block. They were placed outside the vacuum casing about 1 meter from the target on either side of the pipe through which the beam arrived. The target chamber was electrically insulated and was used as a Faraday cage to monitor the beam current. A current integrating device was used which allowed counts from the BF_3 counters to be registered on a scaler until a dose of 500 microcoulombs had been accumulated. Each experimental point on the graphs of Fig. 3 is the total number of neutrons counted for 500 microcoulombs of protons. Since this number is roughly 1.5×10^4, the standard deviation on each point is a little less than 1%.

To avoid stray neutrons from sources other than the Cu crystal, the collimating aperture and the portion of target holder that faced the beam were made of tantalum, which has a low cross section for neutron production. In order to check firstly that these were effective and secondly that they did not introduce any angular effects, a thick polycrystalline Cu target was placed on the holder and counts taken with various proton energies between 2 and 3 MeV. The neutron count was found to follow the expected activation curve for Cu with a threshold at 2.1 MeV. With this polycrystalline target still in place, and a proton energy of 2.8 MeV, the crystal holder was next rotated

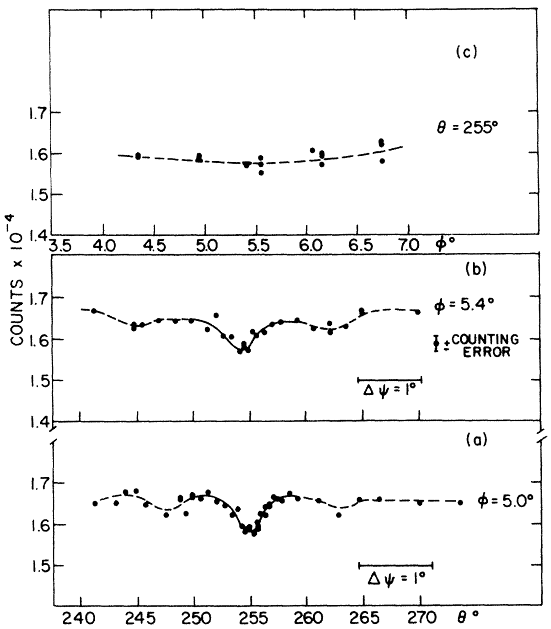

FIG. 3. Number of neutrons counted for 500 microcoulombs of protons, versus angle. The horizontal bars in (c) are from the minima of (a) and (b). Solid and dashed lines are used to indicate regions of greater and lesser certainty.

through a representative range of angles and counts taken to check that there was no spurious effect related to the orientation of the crystal holder.

Finally the crystal was fastened into the holder and with $\varphi = 5.0°$ a series of counts were taken as θ varied through a 30° interval, sweeping the direction of incidence through the [$\bar{1}$11], [1$\bar{1}$0], and [1$\bar{1}$1] planes close to the (110) direction in which they intersect. The change in orientation is illustrated in the stereogram of Fig. 2, and the neutron counts are shown as a function of θ in Fig. 3(a).

Near $\theta = 255°$ a drop in neutron counts is clearly seen. The magnitude of the effect is about 5% which is about five times the standard deviation on a single point and since the minimum value is determined by about ten points, the standard deviation on the minimum value is 0.25%. There is therefore little doubt of the existence of an effect.

The position of the minimum is close enough to the [1$\bar{1}$0] plane on the stereogram for any difference to be attributed to the uncertainty in initial orientation of the crystal. Two subsidiary minima, although less well established, are apparently located on either side of the [1$\bar{1}$0], roughly where the direction of incidence is parallel to [$\bar{1}$11] and [1$\bar{1}$1] planes. In Fig. 3(b) the results are confirmed by a second traverse with $\varphi = 5.4°$. Data not presented in Fig. 3 show minima of comparable magnitude to the [1$\bar{1}$0] where a traverse at $\varphi = 5°$ crosses the [001] plane at $\varphi = 215$ and 275°.

With θ set at 255°, at the minimum for $\varphi = 5°$, a traverse was made by varying φ from 4.35 to 6.75°. The counts are shown in Fig. 3(c) and although there are fewer readings than on the other run, the neutron yield is significantly low throughout the traverse. Since the direction of incidence was swept in the [1$\bar{1}$0] plane, this run proves that the effect is related to planes in the crystal, confirming the earlier finding at low energy[2] and the experiments of Erginsoy, Wegner, and Gibson.[5]

In Figs. 3(a) and 3(b) the change in orientation for given change in θ is shown by a horizontal bar. From this one can see that the angular half-width of the minimum at half-intensity is about 0.15°.

The proton beam current was about 3 μA throughout the experiment and the resultant power input of about 8 watts caused a rise in temperature of the target. Although the temperature was not measured during the run an estimate made at the end of the experiment suggested a value between 200 and 300°C.

In the calculations in reference 2, a screened Coulomb potential was assumed for the ion-atom interaction. The impulse approximation was used to calculate the momentum transfer to the atoms that form the channel wall, and from this the force was calculated which acts to restore the ion to the center of the channel. The channel behavior can be effectively described by a pseudopotential $V(y)$, transverse to the channel axis, with y the displacement from the axis. The transverse motion is then oscillatory, leading to a wavelike trajectory characterized by a maximum displacement or amplitude, y_0, and an angle ψ_0 at which it crosses the central axis. These are related by equating maximum kinetic energy of transverse motion $\psi_0^2 E_1$ to the pseudopotential $V(y_0)$, giving

$$\psi_0 = [V(y_0)/E_1]^{1/2}. \quad (1)$$

Performing this calculation for protons with $E_1 = 2.5$ MeV in the $\langle 110 \rangle$ channel of Cu, one finds that when $\psi_0 = 0.15°$ (the experimentally observed half-width), $y_0 = 0.79$ Å. Since the row of atoms forming the channel wall is at a distance $b = 1.28$ Å from the axis, the proton does not approach closer than $(b-y_0) = 0.49$ Å to the row.

These relative magnetudes show that the rows cannot confine the trajectory to oscillate about a single axis, and for incidence close to ⟨110⟩ rows it must wander, in the Oyz plane, in the manner described by Robinson and Oen.[6] Other trajectories may still be contained between pairs of planes, however, and it is instructive to consider the transition from interplanar to inter-row channeling. The distance traveled forward during reflection from a channel wall is roughly y_0/ψ_0 (reference 2) and in this case is about 400 Å. Suppose the interplanar trajectory makes an angle α with the nearest low-index row in the reflecting plane. If the rows are separated by a, then during reflection the trajectory crosses $n = y_0\alpha/a\psi_0$ rows. The idea of reflection by planes breaks down unless $n \gg 1$. Then one requires that $\alpha \gg a\psi_0/y_0$, and since $a_0/y_0 \sim 1$ for low-index rows, this becomes $\alpha \gg \psi_0$.

In the experiment above, the {100} and {111} effects are certainly interplanar; on the inner sweep across the {110} plane with $\varphi = 5°$, some of the extreme trajectories could perhaps be wandering between ⟨110⟩ rows.

The above treatment neglects lattice vibration and assumes all atoms to lie at their mean positions. This should be a good approximation provided that vibration amplitudes are much less than $(b-y_0)$. The value of $(b-y_0)$ above was calculated for a row; for a close-packed plane the weaker pseudopotential will reduce $(b-y_0)$ to about 0.3 Å. According to the Einstein model for Cu at 250°C the rms displacement of atoms from a plane is ~0.1 Å and the probability of an amplitude greater than 0.3 Å is 10^{-3}. The model should thus be a fair approximation, even for planes, but modification to include lattice vibration is clearly desirable.

The magnitude of the effect on both nuclear reaction and energy loss[3,5] shows that not all trajectories are channeled. There can only be a fraction of the order $y_0/(b-y_0)$ accepted from a uniform flux on geometrical grounds, but the fractional effect is less than this. One possibility is that the initially channeled trajectory is perturbed by collisions with atoms having larger vibration displacements than average, making it revert to the normal trajectory after a shorter distance than that required either to fall below threshold or to emerge from the thin crystal. Such collisions probably set the angular limit on the effect but further investigation will be required.

Measurements of nuclear charged-particle cross section must clearly be dependent on the crystalline nature of the target. In the case above, the angular width of the interplanar channel is apparently of the order of 10^{-2} radian. With the 13 principal planes of a cubic crystal, there is roughly a 6.5% probability of a randomly directed proton beam causing a reaction rate 5% too low. Thus, if one compared a liquid target with a randomly oriented polycrystalline target, assuming the liquid to have a reaction rate similar to that on the plateau between minima in Fig. 3, the liquid should give a 0.3% higher yield. Any other structural phase change should also affect the reaction rate. Equation (1) suggests that the effect should be greatest at low energies. In addition, where one has a threshold energy, the percentage effect should increase as the incident energy approaches threshold and the particle has a shorter distance to travel before becoming ineffective. This has yet to be established.

The assumption has often been made that a solid solution, or chemical compound, containing the reactive nucleus will behave in the same nuclear manner as the pure element, if one introduces a simple dilution factor. This cannot be strictly true as the channeling process and the siting of the reactive nuclei relative to channels must be accounted for.

It has already been shown[2] that radiation damage reduces the channeling effect, presumably because channels are blocked by displaced atoms. Since such damage accumulates in a more disruptive form at lower temperatures, one should expect nuclear reaction rates to increase with decreasing temperature and with increasing irradiation dose. There may also be an effect of lattice vibration, increasing the reaction rate as the temperature is raised. This should not be very important until the temperature exceeds the Debye temperature and the thermal contribution compares with the zero-point motion.

It appears that there is a fruitful field for further study.

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PHOTOIONIZATION OF THE 4*d* ELECTRONS IN XENON*

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Photoionization in the outer electrons of the rare gases has received a great deal of attention in the past few years with the advent of improved technology in the vacuum ultraviolet. In particular the photoionization of the outer electrons in xenon has been studied extensively.[1] On the other hand, comparatively little is known about the absorption characteristics of inner electrons of high orbital angular momentum. The measurements we are about to describe are a study of the photoionization properties of the 4*d* electrons in xenon. In this note we want to emphasize a feature of the photoionization cross section that is unusual. Instead of the orthodox hydrogenic form one assumes for inner-shell photoionization[2] in elements of large Z, we find that the photoionization cross section of the 4*d* electrons in xenon rises to a prominent peak about 32 eV above threshold and drops rapidly thereafter. The cross section at the peak is almost a full order of magnitude greater than the cross section at threshold (the $N_{4,5}$ edges are the ionization threshold for the 4*d* electrons).

The measurements were carried out on a grazing-incidence spectrograph in a wavelength region extending from 80 to 275 Å. The apparatus has been described elsewhere[3] in some detail, but some of the more important features will be noted here. Several combinations of detectors and sources were utilized. In the first case the source was a condensed spark discharge in a glass capillary and the detector was Ilford Q-1 photographic plates. This source consisted of discrete lines from highly ionized atoms, superimposed on a weak continuum. In the second case the source consisted of the characteristic radiation from the beryllium K emission band and the aluminum $L_{2,3}$ emission band. The wavelengths of Be K and Al $L_{2,3}$ band heads fall at 110 and 170 Å, respectively. The Geiger counter detector was used in the scan mode in this case but was not located on the Rowland circle. The definition of images suffered in this mounting. This defect and the presence of wide slits on the counter were responsible for our inability to use the $L_{2,3}$ emission band to investigate the discrete structure in the vicinity of the $N_{4,5}$ edge of xenon.

The key to the experiment lay in a cell in which the gas was confined. A detailed description of the cell and gas-handling techniques can be found in reference 3. The light path was about one centimeter and the gas pressure in the cell ranged from 2.0 Torr to 15 Torr.

The results obtained by the photographic technique represent an average of 18 runs. However, because of the large change in cross section, only a small spectral range could be covered in each run so that the cross section at each spectral line was observed an average of four times.

By observing the deviations from a smooth