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Molecular-Ion Effects in Backscattering from Aligned Silicon Crystals*

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We have observed the proton spectra from equal-velocity beams of H^+, H_2^+, and H_3^+ incident on aligned silicon crystals. Data for both axial and planar channeling show that the dechanneling is successively greater for the H_2^+ and H_3^+ ions than for protons. Since the depth at which dechanneling occured can be deduced from the energy spectrum of the backscattered protons, the comparison of the proton spectrum from molecular-ion bombardment with that produced by incident protons allows us to study the history of the motion in the crystal of the interacting protons from a given molecular ion.

I. INTRODUCTION

Though molecular binding energies lie in the few-eV range, it has become clear that molecular-structure effects can influence processes that take place at much higher energies. As demonstrated in the recent work of Bacher *et al.* there are two main molecular phenomena that cause this influence: the zero-point vibrational energy of the molecule, and the Coulomb interaction of the atoms in the molecule when some of the binding electrons are removed.[1] In the work of Bacher *et al.* these effects produced a significant change in the line shape of a resonance in a nuclear reaction even though the energy of the molecular hydrogen ions involved was 14 MeV/proton.

The dominant effect in these data was produced by the strong Coulomb interaction of the protons. This process could be untangled from the zero-point contribution because the former depends on the target thickness while the latter does not. This dependence arises in the following way: The binding electrons are quickly stripped away by large-angle collisions with other electrons in the target—then the protons from the molecular ion are accelerated away from one another by their Coulomb interaction. In the experiment of Bacher *et al.* the target was sufficiently thin (~ 500 Å) that only a small fraction of the Coulomb energy could be converted to an increase in proton velocity while the protons were in the target (~ 10^{-15} sec). Thus, the contribution to the velocity from the molecular breakup was dependent upon the time the proton remained in the target, i.e., upon the thickness of the target.

The change in the energy of each proton due to molecular effects is

$$\Delta E = \tfrac{1}{2} m \left| \vec{v}_0 + \vec{v}_m \right| - \tfrac{1}{2} m v_0^2,$$

where m is the proton mass, $\vec{v}_0$ is its original velocity, and $\vec{v}_m$ is the velocity coming from the zero-point vibration of the molecule and the Coulomb interaction. Since the latter is time dependent, $\vec{v}_m$ and thus ΔE will also depend on the time.

It is possible to view the analysis of Bacher *et al.* as studying the time history of the breakup of the molecular ion in the target. However, because the molecular effects made the nuclear resonance too broad to be observed for targets thicker than about 700 Å, Bacher *et al.* were limited to studying the breakup only for approximately 10^{-15} sec. In addition, since the effect was folded into the natural shape of the resonance and since there was a strong nonresonant background, the predicted time dependence could only be checked crudely.

Our initial idea was that because the size of a hydrogen molecular ion is slightly smaller than the spacing in the silicon lattice, the channeling process could be used to keep the protons from an individual molecular ion together in the same or adjacent channels so that the breakup process could be studied over a longer interval. This could not

be done in an amorphous solid, because the multiple scattering of the protons by the target electrons would tend to quickly separate them. In addition, by using the channeling process we would be free to change the bombarding energy–Bacher *et al.* were necessarily tied to the energy of the nuclear resonance.

In a preliminary report we have shown that there are indeed significant effects on the channeling process due to the nature of the molecular ion used for bombardment.[2] Similar results have also been obtained by Eisen and Uggerhøj using flux peaking.[3] Even more striking are the data of Poizat and Remillieux, in which it is shown that approximately 10^{-6} of the H_2^+ ions channeled through thin gold crystals (800 Å) emerge as H_2^+ ions.[4] This result tends to support the basic premise upon which our work was initiated: that the protons from a given ion remain correlated because of the channeling process.

In the following sections we shall show samples of our results for the bombardment of aligned silicon crystals with beams of H^+, H_2^+, and H_3^+ and discuss how the data may be used to study the molecular-ion breakup.

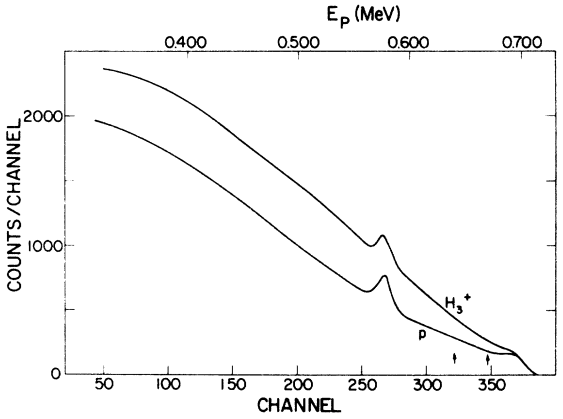

FIG. 2. Proton energy spectra are shown for the backscattering from an aligned ⟨111⟩ silicon crystal. These curves correspond to the minimum of the tilt scan shown in Fig. 1. The arrows show the range of energies summed for the data presented in Figs. 1 and 3. The peak near channel 270 is due to a thin layer of carbon that builds up on the surface of the crystals during bombardment. The plateau at the high-energy end of the proton spectrum is the random scattering yield from near the surface of the target (refer to Sec. IV).

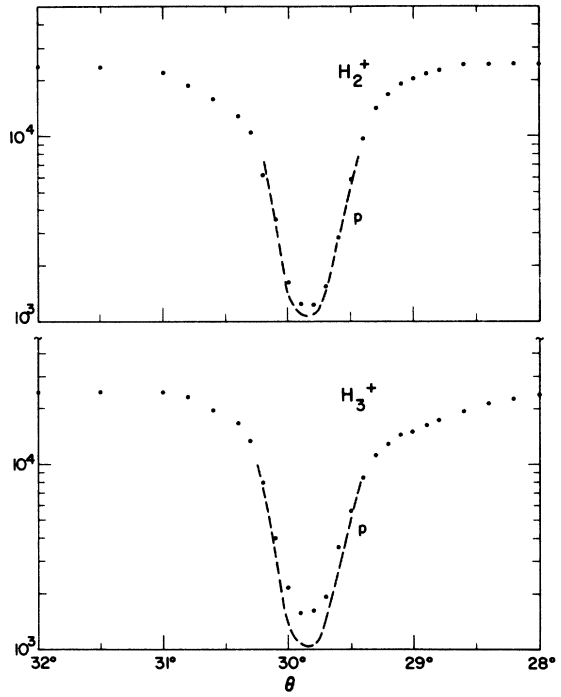

FIG. 1. Comparison of tilt scans through the ⟨111⟩ axial channel on silicon for various molecular-hydrogen-ion species. The vertical scale gives the actual number of counts recorded. The dasned curves correspond to a smooth curve drawn through the data for incident 0.8-MeV protons. The upper half of the figure shows the points for 1.6-MeV incident H_2^+ ions; in the lower half are the points corresponding to 2.4-MeV H_3^+ ions. (Statistical errors at each point are less than the size of the dots.)

II. EXPERIMENT

In the work described here we have used beams of H^+, H_2^+, and H_3^+ having energies of 0.80, 1.60, and 2.40 MeV, respectively. The energies were set to within a few keV using a 90° magnetic analyzer, and the beam was collimated to a half-width angle of 0.03°. The beam current was held to approximately 15 nA to minimize damage to the crystals.

The silicon crystals used were Czochralski grown and were mechanically and chemically polished. They were mounted on a goniometer which allowed the crystals to be rotated about two axes. The backscattered protons were detected with a silicon, surface-barrier detector at an angle of 165° to the incident beam. Each species of molecular ion was directed onto a clean, undamaged spot on the crystal.

Figure 1 shows tilt scans through the ⟨111⟩ axial channel. The procedure was to locate the axial channel and then to scan through the axis by changing the tilt (i.e., longitudinal) angle along a great circle path chosen to lie away from major planar channels. The proton yield shown was taken from the region between the arrows in the proton energy spectrum shown in Fig. 2. This energy region corresponds to scatterings taking place at depths in the crystal between 0.32 and 0.67 μm. (This depth was computed for the minimum in the tilt

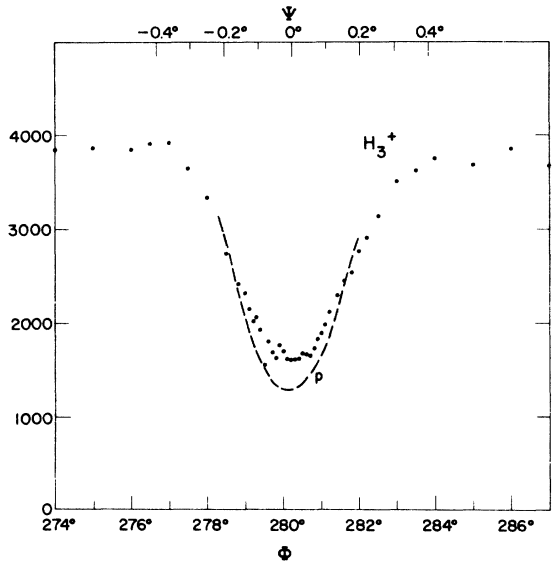

FIG. 3. Comparison of azimuthal scans across the (110) planar channel for incident H_3^+ ions and protons. The vertical scale gives the actual number of counts recorded. The dashed curve is for incident 0.8-MeV protons; the points are for 2.4-MeV H_3^+ ions. Φ is the azimuthal angle reading of the goniometer. The tilt angle was 6.5° for these measurements; the calculated internal azimuthal angle (Ψ) is shown across the top of the figure.

scan; the method of calculation of the energy loss will be discussed in Sec. IV.)

In Fig. 1 the points are shown for incident H_2^+ and H_3^+ ions; the tilt scan for incident protons is shown by a dashed curve for clarity. It is obvious that there is an enhancement of the dechanneling for the molecular ions relative to protons—with H_3^+ showing a much larger effect than H_2^+. This effect is reproduced for the protons backscattered from all depths on both the ⟨111⟩ and ⟨110⟩ axial channels.

Figure 3 shows a scan across the (110) planar channel after the polar angle had been shifted by 6.5° from the ⟨111⟩ direction. Only the results for H_3^+ are shown because the difference between the H_2^+ data and the proton curve are barely outside our statistical accuracy. For H_3^+ the magnitude of the effect is much less than for axial channeling, but is still appreciable.

The proton energy spectra for incident protons (lower curve) and for H_3^+ (upper curve) are shown in Fig. 2, corresponding to the bottom of the dip in Fig. 1 (aligned). The peak near channel 270 corresponds to the buildup of a thin carbon layer ($\sim 10^{16}$ atoms/cm^2) on the surface of the crystal under bombardment. The energy scale shown was calibrated by scattering protons from targets of several different elements. The curves in Fig. 2 confirm that there is a strong enhancement in the dechanneling when molecular ions are used and that the effect extends deep into the crystal.

In Sec. III we shall show how the molecular breakup is responsible for the effects observed, and in Sec. V we shall attempt to extract from the data information about the history of the interaction of the protons from a molecular ion as they move down the channel.

III. A MODEL OF THE BREAKUP PROCESS

In order to estimate the effect produced by the breakup of a molecular ion on the channeling process, we must first have an *accurate* description of proton channeling. However, this does not imply that a *detailed* description of the process be obtained; it will be shown that we need only identify the variables that are involved. Following the usual approach, we shall assume that the component of the momentum transverse to the channel axis is sufficient to completely specify the dependence of the proton backscattering yield on the tilt angle. Thus,

$$\mathcal{Y}_p(\theta) = \mathcal{Y}_p(p_0 \sin\theta) ,$$

where p_0 refers to the momentum of the protons in the beam. Figure 4 defines the coordinate system used: The z axis lies along the channel axis; the x axis lies across a planar channel.

We may now use the proton results in our description of the molecular-ion effects. Initially each proton in the molecular ion has a momentum $\vec{p}_0$—the breakup process adds a component $\vec{p}_m$.

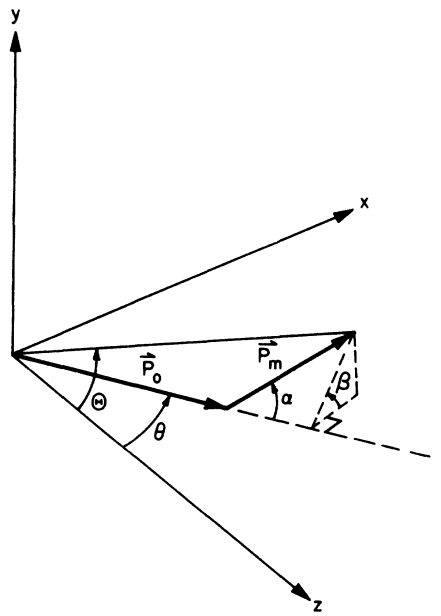

FIG. 4. Coordinate system used in describing the channeling of a molecular ion. The axes and angles shown are defined in Sec. III of the text.

Thus, the resultant momentum is $\vec{p}_0+\vec{p}_m$. We now need to find the component of this momentum that is transverse to the channel axis in terms of the original angle θ. Remembering that $p_m \ll p_0$ and $\theta \ll 1$, we obtain

$$p_{\text{trans}} = p_0 \sin\Theta \text{ for axial channeling,}$$

where

$$|\Theta| = (\theta^2 + 2\eta\theta \sin\alpha \cos\beta + \eta^2 \sin^2\alpha)^{1/2} .$$

The sign of Θ is the same as that of $\theta + \eta\sin\alpha\cos\beta$. In these expressions $\eta = p_m/p_0$, and α and β are the polar and azimuthal angles of the breakup with respect to the original direction of motion.

The backscattering yield for an incident molecular-ion beam at a tilt angle θ is, therefore,

$$Y_m^{\text{axial}}(\theta) = \frac{1}{4\pi}\int_0^{2\pi} d\beta \int_0^{\pi} \sin\alpha \, d\alpha \, N(\alpha, \beta)\, y_p^{\text{axial}}(\Theta),$$

where $y_p(\Theta)$ is the yield for incident protons at a tilt angle Θ, and $N(\alpha, \beta)$ allows for a possible alignment of the molecular ion with respect to its direction of motion. [If its orientation is random, then $N(\alpha, \beta) = 1.$] In general, both p_m and $N(\alpha, \beta)$ must be assumed to be time dependent, i.e., dependent on the depth in the crystal.

For planar channeling only the component of $\vec{p}_0 + \vec{p}_m$ in the x direction is needed:

$$Y_m^{\text{planar}}(\psi) = \frac{1}{4\pi}\int_0^{2\pi} d\beta \int_0^{\pi} \sin\alpha \, d\alpha \, N(\alpha, \beta)\, y_p^{\text{planar}}(\Psi),$$

where $\Psi = \psi + \eta\sin\alpha\cos\beta$. (In this case, ψ is the internal azimuthal angle.)

The technique just described has the obvious advantage that the details of the channeling mechanism as well as resolution effects do not have to be described explicitly—they are implicitly contained in the proton-backscattering-yield curve.[5] It is clear that in arriving at this result we have made a restrictive assumption in neglecting the explicit time dependence of the dechanneling process. To include this effect obviously demands the inclusion of the dynamics of the channeled particles in the treatment. In the absence of such detailed calculations, the approximation described here is intended to yield only a qualitative understanding of the data.

In order to show an example of how the molecular contributions can strongly modify the channeling process, let us consider the case of a Coulomb explosion of the H_2^+ ion. (The rotational energy of the molecular ion can be neglected; however, one must look carefully into possible contributions from vibrational states. The zero-point motion turns out to be small compared to the Coulomb interaction. Walters *et al.* have estimated the population of excited vibrational levels in the molecular-ion beam—giving a contribution of less than 1 eV/proton.[6] Since in the case described here all these contributions are small compared to the Coulomb energy, they will be neglected.) The terminal energy associated with the explosion is 6.8 eV/proton (16.5 eV/proton for H_3^+). Since 2/3 of this energy is achieved before a depth of 600 Å is reached, we shall use the full terminal energy in our calculations. Screening of the Coulomb interaction of the protons within a solid is not expected to appreciably modify this terminal energy. This results because when the protons are close together, in the short time that they remain near a given electron it cannot be displaced enough to modify the protons' Coulomb interaction.[1] When the protons are far apart most of their Coulomb energy has already been converted to kinetic energy, so that the presence of screening at large distances is of little consequence in computing the terminal energy.

As given earlier in this section,

$$\Theta^2 = \theta^2 + 2\eta\theta \sin\alpha \cos\beta + \eta^2 \sin^2\alpha.$$

For H_2^+ at $\theta = 0.5°$,

$$\theta^2 = 7\times10^{-5},$$

$$2\eta\theta = 5\times10^{-5},$$

and

$$\eta^2 = 0.7\times10^{-5}.$$

Thus, since all the terms are of comparable size, appreciable molecular effects are to be expected. (For the case of maximum contribution, $\alpha = 90°$ and $\beta = 0°$, the resulting value is $\Theta = 0.65°$.)

Because the transverse motion of the protons from a molecular ion is bounded by the channel, the Coulomb force between them serves mainly to increase their separation. A crude diagram of this effect is shown in Fig. 5 for the H_2^+ ion. One immediately sees that this may be interpreted as an effective "alignment" of the molecular constituents. Thus, though $N(\alpha, \beta)$ is probably equal to unity at the surface of the crystal, it becomes peaked toward $\alpha = 0°$ and $\alpha = 180°$ as the depth increases.

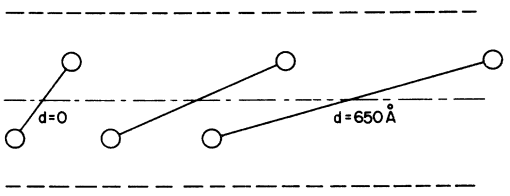

FIG. 5. Simplified diagram of the "alignment" of the protons from an H_2^+ ion by the channeling process. The heavy dashed lines are the channel boundaries; the circles are the protons in an H_2^+ ion. The quantity d refers to the depth in the crystal. The protons are pushed apart by their Coulomb repulsion, but since the transverse motion is bounded by the channel, the angle between the axis of the "molecule" and the channel axis decreases with increasing depth.

If we neglect a possible difference in the energy loss suffered by each proton, the separation of the pair from their Coulomb repulsion may be calculated. Combining this separation distance with the fixed width of the channel thus provides a crude estimate of the effective "alignment"; e.g., by a depth of 5000 Å the particles could be within a half-angle of 4°.

In Sec. IV we shall use the techniques we have just described to deduce the history of the ion breakup. Since the Coulomb explosion reaches its terminal stage relatively quickly, the time dependence of the acceleration process can be ignored and the terminal energy will be used in all calculations. Thus, the only time-dependent quantity considered is $N(\alpha, \beta)$.

IV. ANALYSIS

The energy spectrum of the backscattered protons will yield the depth (or time) at which a proton left the channel if we can obtain the connection between the particle energy and the depth. Obviously, the key ingredient in this calculation is the knowledge of the energy loss. We have taken the energy loss for the channeled protons (aligned case) from the data of Della Mea *et al.*[7]; the energy loss of the protons after backscattering (random case) was taken from Williamson *et al.*[8] The $(dE/dx)_{\text{aligned}}$ curve for the channeled protons was integrated from the surface to the depth at which backscattering occurred; the energy was then computed for scattering from silicon at 165°; then the $(dE/dx)_{\text{random}}$ curve was integrated back to the surface. This procedure allows the dependence of the observed energy on the backscattering depth to be determined, if we assume that backscattering occurs rapidly after the proton is dechanneled. This assumption is, of course, expected to be well justified in the present experimental arrangement. We also have been forced to assume (in the absence of other data) that the energy loss is not affected appreciably by the interaction of the pair of protons.

The small plateau at the high-energy end of the energy spectrum shown in Fig. 2 represents the "random fraction" of the incident protons. These are protons that scatter from the ends of the rows of the crystal lattice or the carbon or SiO amorphous layers at the surface of the target and are immediately dechanneled. We will, therefore, remove this fraction from the data in making our analysis. This fraction amounts to approximately 4% of the random backscattering yield and was determined from the plateau of the proton energy spectrum shown in Fig. 2.

To show the molecular effect we have plotted in Fig. 6 the ratio

$$\frac{Y_m(d) - Y_{p,\text{random}}}{Y_p(d) - Y_{p,\text{random}}}$$

versus the depth d at which backscattering occurred. The Y_m and Y_p are the yields versus backscattering depth corresponding to the minimum of the axial tilt scan for molecular ions and protons, respectively. The $Y_{p,\text{random}}$ is the yield associated with the random fraction of the incident protons described in the previous paragraph.

The first 5000 Å is characterized by excess dechanneling, which is probably explained partially by the fixed spatial configuration of the molecular ion that causes one proton to have a higher probability to hit the end of a row of the lattice if the other proton channels. (One should keep in mind that though the ratio is large, the number of particles involved is small in this part of the energy spectrum.)

The open points shown correspond to calculations

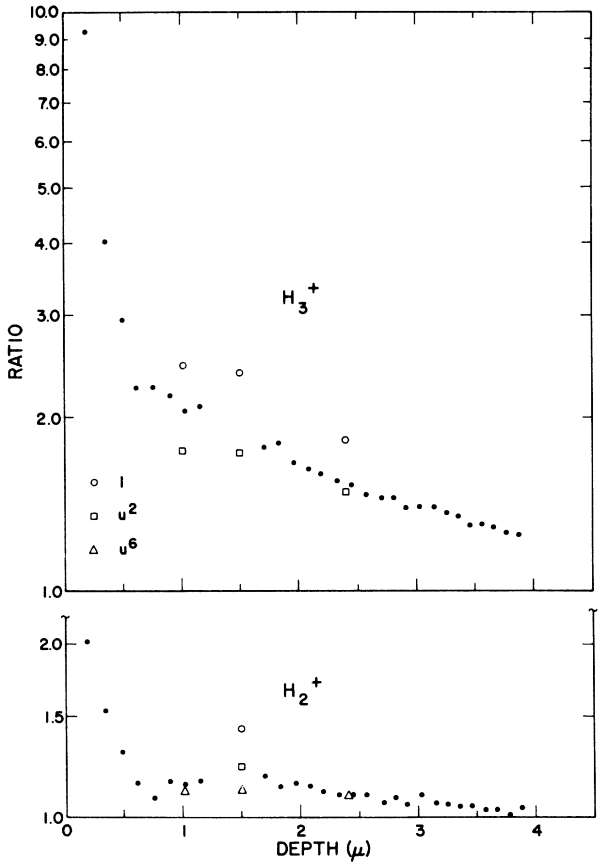

FIG. 6. Ratio of channeling yield for H_3^+ and H_2^+ to that for protons at various depths (in μm) in the aligned ⟨111⟩ silicon crystal. The solid points are taken from the data; the open points are calculated for various forms of $N(\alpha, \beta)$. Refer to Sec. IV of the text for a detailed discussion.

using the techniques of Sec. III for various choices of $N(\alpha, \beta)$. We have shown three choices: $N(\alpha, \beta) = 1$, $N(\alpha, \beta) = 3\cos^2\alpha$, and $N(\alpha, \beta) = 7\cos^6\alpha$, which are referred to in Fig. 6 as 1, u^2, and u^6, respectively.

For H_3^+ at depths greater than 0.5 μm the data start off like $N(\alpha, \beta) = 1$ but by the time a depth of 3 μm has been reached u^2 is a better description. The interaction of the three protons in the H_3^+ (an equilateral triangle configuration) to give a slight alignment with depth is reasonable, but a quantitative estimate would be rather difficult.

For H_2^+ the data are well described by the u^6 alignment—this corresponds to an average angular opening of 8°, which is actually larger than the estimate of 4° given in Sec. III. As one would expect, the simple H_2^+ configuration is easier to align than that of the H_3^+ ion. If other effects are considered, it does not seem surprising that the molecular ion cannot become arbitrarily well aligned with increasing depth, but should approach some equilibrium form of $N(\alpha, \beta)$ that is determined by the thermal motion of the lattice and by the screening of the Coulomb field of the two protons at large separations.

V. CONCLUSION

The results presented in this paper have shown that the interaction of the constituents of a molecular hydrogen ion within a silicon crystal produce appreciable effects in both the energy spectra of the backscattered protons and in the shape of the minimum in the axial and planar scans. Such collective effects should also appear when one observes the energy loss of protons from molecular-ion bombardment after transmission through an aligned crystal.

In a more speculative vein, one might also expect to see collective (or perhaps even coherent) effects due to the interaction of the correlated protons with the crystalline lattice. Such effects could arise because of the short time interval (10^{-15} to 10^{-17} sec) between the collisions of the protons with a given lattice atom and the periodic nature of the motion of the channeled particles. Though these effects may be observable in high-resolution measurements of the energy losses of the channeled particles, a more sensitive scheme would probably involve the detection of x rays from the lattice atoms—probably in coincidence with the transmitted protons.

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