detection is being carried out. In any event, we would expect in sufficiently pure material to see structure in the ultrasonic attenuation whenever an acoustic wave of wavelength $\lambda \sim d$ propagates in a direction perpendicular to the magnetic field. The wavelength at which such structure occurs is a direct measure of the distance between flux lines for a given magnetic field.

If the effects predicted above are accessible to experiment, ultrasonic attenuation could become a very valuable tool for probing the detailed characteristics of the mixed state of type-II superconducitivity.

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CRYSTAL STRUCTURE OF SOLID DEUTERIUM FROM NEUTRON-DIFFRACTION STUDIES*

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The earliest and perhaps the most definitive investigation of the crystal structure of any of the solid isotopic hydrogens is that of Keesom, De Smedt, and Mooy.[1] The observed x-ray Debye-Scherrer patterns of more or less randomly oriented crystallites of solid parahydrogen ($J=0$, ground rotational state) at liquid-helium temperatures were indexed on the basis of a close-packed hexagonal structure; $a=3.75$ Å, $c/a=1.633$. Several other less direct experiments have lent support to this close-packed structure for both isotopic species, hydrogen and deuterium, whether in the ground rotational state ($J=0$) or, in part, rotationally excited (ortho-para mixtures). An analysis of the heat-capacity date of solid hydrogen[2,3] and deuterium[4,5] above the λ transition ("high-temperature tail" of anomaly) is in accord with the close-packed assignment for their structures. Furthermore, the interpretation of the infrared absorption spectra[6,7] of the solid isotopes, above the λ transition, requires a hexagonal close packing to account for both the observed bands and their intensities.

Reports of recent investigations of the crystal structure of the solid hydrogens by neutron-diffraction, x-ray-diffraction, and electron-diffraction techniques have tended to confuse the situation. The initial x-ray and neutron-diffraction results of Kogan and collaborators[8] were interpreted on the basis of a body-centered tetragonal structure for the two solid isotopic species, hydrogen and deuterium. However, their more recent x-ray studies[9] reportedly confirm the hexagonal close-packed structure of Keesom, De Smedt, and Mooy[1] for hydrogen and suggest a hexagonal structure, $c/a=1.67$ for solid deuterium. The electron diffraction studies of Curzon and Pawlowicz[10] do not support the above assignment for solid deuterium. The diffraction patterns have been indexed on the basis of a close-packed face-centered cubic lattice.

It is clear from the early work of Keesom, De Smedt, and Mooy[1] and the more recent structure studies[8-10] that a major problem in powder diffraction studies of the solid hydrogens is the preparation of randomly oriented crystallite samples. We have recently succeeded in preparing such solid samples in the case of "ortho"-deuterium (97.8% ortho, $J=0$; 2.2% para, $J=1$) and normal deuterium ($66\frac{2}{3}$% ortho, $33\frac{1}{3}$% para). These have yielded reproducible neutron-diffraction data, with respect to both line positions and intensities from which an unequivocal assignment of the structure of these solids can be made.

The solid deuterium samples were prepared by rapid freezing of the liquid in a thin-walled aluminum cell, 1.6 in. long and 0.56 in. in diameter. To ensure formation of a randomly oriented powder the cell was filled with a porous matrix, fabricated from an aluminum-magnesium alloy[11] (90% Al, 10% Mg), with a void volume of nearly 67%. About 0.2 mole of liquid deuterium was condensed in this cell. The rapid freeze was accomplished by placing the liquid deuterium in thermal contact with a vessel containing solid hydrogen maintained at 13°K. To test whether random orientation of crystallites had been achieved, the angular dependence (rotation about the axis of the cylindrical cell which is maintained normal to the neutron beam) of the most intense reflection, $2\theta \approx 20°$, was measured. In the experiments reported below the maximum variation of intensity, on a 360° rotation, was less than ±20%. Furthermore, the line positions and intensities were reproducible on successive melting and recrystallization of a given deuterium sample.

The neutron-diffraction data for a sample of "ortho"-deuterium at 13°K are shown in Fig. 1. (The background counts, n_B, due to the empty cell have been subtracted from the total measured counts, n_T.) The counting time for each of the points, shown in Fig. 1, was approximately 12 minutes, and the cryostat was rotated about the cylindrical axis of the sample cell at a rate of two revolutions per minute. The detector used in these experiments was equipped with a Soller collimator having an azimuthal angular aperture of approximately 0.9° and a height of two inches at a distance eight inches from the center of the sample. The wavelength of the incident neutrons was 0.959 Å.

It is not possible to index the observed reflections as a single phase on the basis of either a cubic or tetragonal lattice. If the structure is no more complex than hexagonal, the complete assignment of indices, shown in Fig. 1,

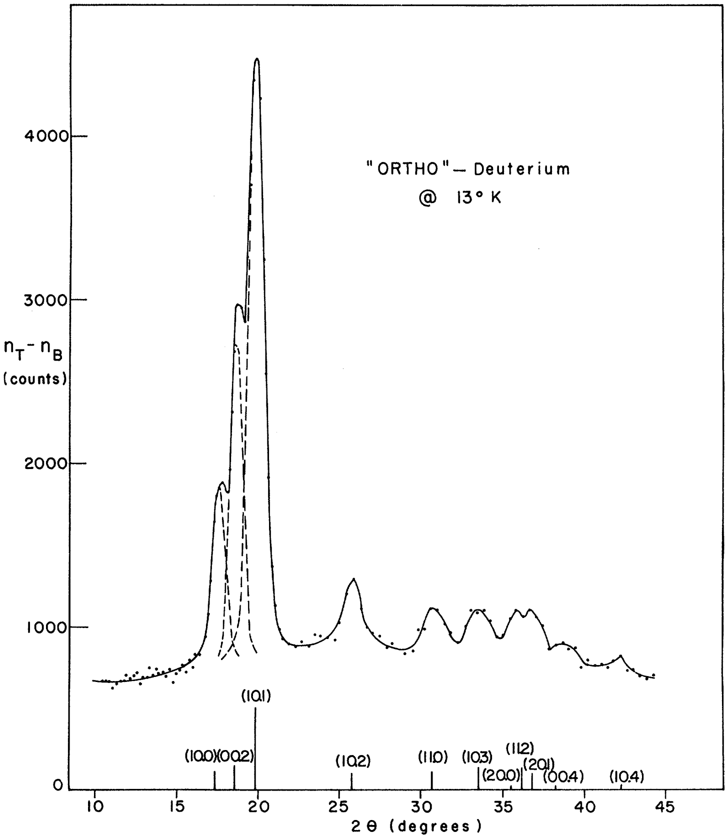

FIG. 1. Points on full line: n_T-n_B versus (Bragg) 2θ at 13°K. Dashed line: resolution of lines used for calculation of lattice spacings. Vertical lines: angle position computed from cell, $a = 3.62$ Å, $c/a = 1.61$; line height is relative measure of integrated intensity computed from molecular cross section of "ortho"-deuterium.

can be made as a close-packed structure containing two molecules per unit cell. The cell dimensions from a least-square calculation utilizing eight of the observed reflections are $a = 3.63$ Å and $c/a = 1.61$. It is interesting to note that the intensity distribution is very similar to that of other close-packed hexagonal structures, e.g. magnesium, where the reflections $(00l)$ for l odd and the (111) are missing. Since $(00l)$ reflect for l even, one-half the scatterers are related to the other half by a vector having a z component $z/2$, or very nearly so. The only two-site hexagonal cell fulfilling this requirement and the requirement that the (111) reflection vanish is $(0,0,0)$, $(\frac{1}{3},\frac{2}{3},\frac{1}{2})$, of space group $D_{6h}{}^4$, i.e., hexagonal close-packed. There are two possibilities under this space group: (a) one in which the molecular axes are correlated in the cell, i.e., the individual atoms have specifiable sites, $D_{6h}{}^4$, case[12] $4f$ (site symmetry $3m$); (b) the uncorrelated case, compatible with random orientation of the molecular axes or free rotation, $D_{6h}{}^4$, case[12] $2c$ (average site symmetry $\bar{6}m2$), the highest site symmetry possible for hexagonal close packing. Until a detailed analysis is made of the integrated intensities, it is not possible to distinguish between these two cases. Support for random orientation of molecules, case (b), can, however, be found in the experimentally observed axial ratio, $c/a = 1.61$, which is very nearly 1.633, as required for close-packed spheres. Furthermore, relative intensities, calculated on the basis of the coherent cross section of molecular deuterium,[13] shown by vertical lines perpendicular to the abscissa in Fig. 1, are in qualitative agreement with the measured integrated intensities.

The molar volume of solid "ortho"-deuterium calculated from the unit cell dimensions is 20.1 cc. This is in excellent agreement with the measured molar volume of the solid,[14] 20.05 cc, interpolated at 13°K.

Neutron-diffraction data have also been obtained for a solid sample of normal deuterium ($66\frac{2}{3}$% ortho, $33\frac{1}{3}$% para) at 13°K. Within experimental error these data are the same as those for the "ortho"-deuterium, both with respect to line positions and intensities. It is therefore concluded that the crystal structures of "ortho" and normal deuterium are the same, at least in the temperature range 13°K to the triple points. Experiments are now under way to investigate the neutron diffraction from solid deuterium highly enriched in para content, both above and below the λ anomaly[4] where cooperative rotational quenching occurs.

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