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a-Si : SHORT RANGE ORDER BY NEUTRON SCATTERING

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Résumé — Cet article décrit une expérience de diffusion de neutrons destinée à déterminer le rôle joué par l'hydrogène dans l'ordre local du silicium amorphe hydrogéné. L'utilisation de la méthode de substitution isotopique nous a permis d'obtenir les trois facteurs de structure partiels à partir desquels ont été calculées les trois fonctions de corrélation de paires correspondantes. La présence de groupements Si-H et Si-H$_2$ a pu être mis en évidence directement. Ces groupements ne perturbent pas sensiblement l'ordre local tétraédrique du silicium amorphe.

Abstract — We report in this paper a neutron scattering study of a-Si:H, undertaken in order to point out the part played by hydrogen in the structural properties. The use of the isotopic substitution method provide us with the three partial structure factors from which have been deduced the three corresponding pair partial correlation functions. Si-H and Si-H$_2$ units have been directly put into light. These hydrogenated units have not produced noticeable distortion of the tetrahedral local order.

I - INTRODUCTION

Hydrogenated amorphous silicon a-Si:H has been the subject matter of a lot of both experimental and theoretical studies due to its semiconducting properties /1/.

Introducing hydrogen reduces the DOS of several orders of magnitude and allows n or p doping of a-Si:H. However, owing to the weak sensitivity of hydrogen to most of scattering experiments, our knowledge of local order still remains very poor. Therefore, in order to try to determine the mechanism of interaction of hydrogen with silicon in a-Si:H we have undertaken a neutron scattering structural study based on the strong interaction between neutrons and protons.

II - ISOTOPIC SUBSTITUTION METHOD

The scattered intensity for a binary mixture, the coherent scattering length of which are $b_1$ and $b_2$ corresponding respectively to concentrations $c_1$ and $c_2$, can be written:

$$S(Q)-1 = (c_1^2 b_1^2 + c_2^2 b_2^2)^{-1} \left( c_1^2 b_1^2 (S_{11}(Q)-1) + c_2^2 b_2^2 (S_{22}(Q)-1) + 2c_1 c_2 b_1 b_2 (S_{12}(Q)-1) \right)$$

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where \( S_{ij} \) represent the partial structure factors (PSF'S) in the Faber-Ziman formalism.

The pair partial correlation functions (PPCP'S) which can be obtained as the Fourier transforms of these PSF'S provide us with the most precise possible information about local order in a disordered system. However the determination of the three PSF'S requires at least three independent equations.

Using neutron scattering this condition could be fulfilled by using three isotopic compositions of one component corresponding to different scattering length. This method has been intensely used during the past 20 years. However as far as hydrogen and deuterium have opposite sign coherent scattering length, the proton is very well suited for a structural study using isotopic substitution method.

**III - EXPERIMENTAL RESULTS**

Our samples were prepared by sputtering at high temperature (400°C) in order to get rid of large defects such as voids (this point has already been tested by SANS on D17 spectrometer at ILL Grenoble /2,9/). Three isotopic concentrations have been used to provide us with a set of total structure factors, out of which the PSF'S could be extracted, H, H, H,6D,4 and D.

Owing the very different scattering lengths of the various samples (mainly due to the hydrogen incoherent cross section), the counting times of the runs have been carefully chosen to provide quite similar statistical error on the coherent part of the scattered intensity from each sample. This choice was possible after a preliminary experiment performed at the reactor Orphée at Saclay.

Data reduction have been performed at the L.L.B. in Saclay. Usual corrections have been applied for absorption, multiple and incoherent scattering. Due to the very high incoherent inelastic scattering from hydrogen, a classical Placzeck correction was quite unreliable. Therefore, correction for inelastic scattering has been made using a procedure due to Chieux et al /3/.

\[
\begin{array}{ccc}
\text{b} & \text{b} & \sigma (10^{-24} \text{cm}^2) \\
\text{Si} & .4149 & 2.17 \\
\text{H} & -.3741 & 81.7 \\
\text{D} & .6674 & 7.6 \\
\end{array}
\]

**Table 1**

Coherent scattering lengths and total cross sections

We give in the table 1 the coherent scattering length (\( b \)) and the total cross sections (\( \sigma \)) for silicon, hydrogen and deuterium /4/. Using the three isotopic compositions given above, we have determined three total structure factors from which PSF'S have been calculated.

As it can be seen on the figure 1, the hydrogen-hydrogen structure factor presents a very large uncertainty due to the part played by the very large hydrogen incoherent cross section (see table 1).

The PPCP'S, \( G_{ij}(r) \) have been obtained by Fourier transform of the
PSF'S:

\[ G_{ij}(r) = 1 + (2\pi^2 \rho)^{-1} \int_0^\infty Q(S_{ij}(Q)-1) \sin(Qr) dQ \quad (2) \]

\( \rho \) is the average number density.

The three PPCF'S are represented in the figure 2, in which we have reported the maxima of the two first distributions of distances.

IV - DISCUSSION

The very well defined position of the two first peaks of the silicon-silicon PPCF, clearly put into light the tetrahedral local arrangement of silicon atoms. The presence of a shoulder located at about 4 Å and the relatively high value of Si-Si PPCF at 4.5 Å indicate that a mixture of staggered and eclipsed arrangement is more probable than a random distribution of dihedral angle.

The location of the two first peaks of the silicon-hydrogen PPCF is very consistent with the existence of Si-H and Si-H\(_2\) units, as far as it respectively corresponds to the H distance to first and second silicon neighbours (see figure 3). This result has been already assumed as an explanation of IR vibration spectra (5), however this neutron determination provide us with the first direct evidence of such distances.
Due to the poor definition of the hydrogen-hydrogen PSF, no reliable structural feature can be extracted from H-H PPCF. However the distance at 2.2 Å should indicate the presence of Si-H₂ units.

![Diagram of Si-H units](image)

Fig. 3. Schematic local order in a-Si:H

V - CONCLUSION

These neutron scattering experiments on a-Si:H have demonstrated that local order of the silicon matrix was based on tetrahedral structure, in spite of the existence of Si-H and Si-H₂ units which could distorted the silicon lattice.

The importance of corrections and of the statistical errors do not allow us to see molecular hydrogen, the existence of which has been assumed by RMN /6/ and confirmed by calorimetric measurements /7-8/. This result could be consistent with an heterogeneous distribution of hydrogen demonstrated by SANS experiments /9/.

More precise measurements in a wider momentum transfert range will be done in a next future, in order to determine the relative proportions of Si-H, Si-H₂ units and molecular hydrogen.

REFERENCES

/1/ Amorphous and Liquid Semiconductors: