PHYSICAL REVIEW B VOLUME 23, NUMBER 6 15 MARCH 1981

Proton spin-lattice relaxation in plasma-deposited amorphous silicon-hydrogen films

Jeffrey A. Reimer* and Robert W. Vaughan†
Division of Chemistry and Chemical Engineering, California Institute of Technology, Pasadena, California 91125

John C. Knights
Xerox Palo Alto Research Center, 3333 Coyote Hill Road, Palo Alto, California 94304

(Received 22 September 1980)

Proton spin-lattice relaxation data are presented for several plasma-deposited amorphous silicon-hydrogen films when (i) homonuclear dipolar interactions are suppressed, (ii) deuterium is isotopically substituted for hydrogen, and (iii) films are annealed. These data are consistent with a model in which proton nuclei are relaxed by hydrogen-containing disorder modes. Analysis of these data shows that the density of disorder modes is ~30% higher in the low-hydrogen-density domain and that more than one hydrogen nucleus is associated with each disorder mode. The behavior of T_1 upon annealing indicates that a small fraction of unpaired spins or "dangling bonds" may be associated with disorder modes. These results suggest that the role of hydrogen in amorphous silicon is more complex than passivation of dangling-bond intrinsic defects.

I. INTRODUCTION

Hydrogenated amorphous silicon (a-Si:H) has attracted a great deal of attention recently because of its remarkable electronic properties *vis-à-vis* pure amorphous silicon. The hydrogenated material,[1] prepared either by glow discharge of silane or reactive sputtering, has a large photoconductivity, efficient photoluminescence, and a very low concentration of unpaired electron spins associated with "dangling bonds." Because the hydrogen apparently passivates[2] the large number of these unpaired spins found in pure amorphous silicon, controlled doping of hydrogenated films has led to the development of p, n, and Schottky-barrier junctions,[3–5] as well as moderately efficient solar cells.[6]

More recently, a variety of proton nuclear magnetic resonance (NMR) data have posed some new questions on the role of hydrogen in both electronic and structural properties of plasma-deposited inorganic films. Linewidth and line-shape results[7] have shown that plasma-deposited a-Si:H is characterized by two domains which differ in the local density of hydrogen. Furthermore, upon annealing[8] hydrogen in the less clustered domain diffuses internally concomitant with the reduction of paramagnetic center density.[9] Proton spin-lattice relaxation measurements in a-Si:H have also been reported recently[10] and were explained by postulating the existence of hydrogen-containing disorder modes that may contribute to the electronic states within the band gap. In fact such disorder modes have been observed directly by us using proton NMR in amorphous boron, carbon, and silicon nitride.[11]

The purpose of this work is to investigate the proton spin-lattice relaxation behavior in a-Si:H in detail in order to ascertain the validity of the disorder-mode relaxation model and to examine the structural details of both the disorder modes and their distribution in the film. An understanding of these details is expected to aid in discerning the relationship between film microstructure and defects which contribute to nonradiative recombination processes and which limit luminescence efficiency. In this paper we present proton spin-lattice relaxation behavior when: (i) homonuclear dipolar interactions are suppressed, (ii) deuterium is isotopically substituted for hydrogen, and (iii) films are annealed under conditions which produce rapid changes in unpaired spin density and luminescence efficiency. The data provide additional evidence for the relaxation model of Carlos and Taylor[10] and yield new information on the distribution of hydrogen-containing disorder modes within the film, the number of hydrogen nuclei within the disorder modes, and the association of dangling-bond paramagnetic centers with the disorder modes.

II. EXPERIMENTAL

Magnetic resonance data were taken with an NMR spectrometer previously described[12] with an operating frequency of 56.4 MHz (1.3 T). Proton spin-lattice relaxation times (T_1) were determined by the inversion recovery method.[13] Proton spin-lattice relaxation times, while homonuclear dipolar interactions were suppressed, were determined by the T_{1y} method.[14] We shall briefly review the T_{1y} technique since it is relatively new and only recently received attention in the literature.[14,18,30]

In order to suppress homonuclear dipolar interactions an eight-pulse cycle,[15]

$$\tau^* - P_x - P_y - 2\tau - P_{-y} - \tau - P_{-x} - 2\tau - P_{-x}$$
$$- \tau - P_y - 2\tau - P_{-y} - \tau - P_x - \tau ,$$

was applied to the spins. Here P_x, etc., denote 90° pulses in the directions x, etc., and τ and 2τ stand for the pulse separations. At the point τ^* in the cycle the magnetization is sampled and stored in a transient reorder. The cycle is repeated 1024 times, continuously, and the magnetization is thus "stroboscopically" observed. The effective rotating-frame Hamiltonians for the spins under this pulse cycle have been calculated previously[15,19,20] via the Magnus expansion approach and yield the homonuclear dipolar interaction as zero to second order. The chemical shift Hamiltonian becomes[15]

$$\bar{\mathcal{H}}_{cs}^{(0)} = \sum_i \frac{\alpha(\Delta\omega + \omega_0 \sigma_{zzi})}{3} (I_{xi} + I_{zi}) ,$$

where α is the chemical shift scaling factor, ω_0 the resonant frequency of the spins, $\Delta\omega$ the amount the applied rf differs from ω_0, and σ_{zz} the chemical shift tensor component for the I spin. This Hamiltonian is thus in the [101] direction and, in geometric terms, the initial magnetization may be thought of as precessing about the [101] direction.

In the T_{1y} experiment a 90^0_x or 90^0_{-x} preparatory pulse is used to place the total thermally induced magnetization parallel to the y direction. While applying this eight-pulse sequence on resonance a phase error of ~2 kHz in the x pulses is introduced, which gives rise to a y Hamiltonian,

$$\bar{\mathcal{H}}_{P}^{(0)} = \frac{2}{t_c} (-\phi_x + \phi_{-x}) I_y ,$$

where t_c is the eight-pulse cycle time and ϕ_x is the phase misadjustment of the x pulses. Under this Hamiltonian the magnetization is forced to precess about the y axis (perpendicular to the [101] direction) and thus "second averages"[19,20] chemical shift and field inhomogeneitites to zero. The small nonfluctuating phase error Hamilitonian does not affect relaxation times[14]; hence, the observed decay is due only to spin-lattice relaxation. However, the effective frequency for relaxation is not the Larmour frequency, as in conventional T_1 measurements; rather, it is given by the cycle time of the eight-pulse sequence. This is analogous to $T_{1\rho}$ "spin-locking" measurements[31] where the effective field is defined by the magnitude of the locking field. For these data a 50-μsec cycle time was used for the eight-pulse sequence, and the 90° pulse widths were less than 2 μsec. The 90^0_x and 90^0_{-x} preparatory pulses were alternately applied, and the data were cosubtracted with 100–200 acquisitions accumulated to improve the signal-to-noise ratio and to eliminate base line artifacts.

The samples were prepared in an rf-diode deposition system, details of which have been published elsewhere.[16] Five samples were prepared that represent a proton content of 31 to 7 at.% by varying the deposition parameters. The hydrogen content was determined by using the integrated proton spin density and the sample weight. The samples were deposited onto 2-in.-diameter aluminum foil substrates in thicknesses of approximately 10 μm, resulting in sample masses of 50 mg after removal of the substrates with a dilute hydrochloric acid etch. The notation for the deposition conditions used in the text are as follows: (5% SiH_4-Ar, 18 W, RT, C) refers to deposition from 5% silane diluted in argon, a rf power (net into matching network) of 18 W, a substrate temperature of 25 °C, and deposition onto the hot rf electrode (cathode), as opposed to the grounded electrode (anode). Some samples were then placed in a quartz NMR tube and isochronally annealed at various temperatures under flowing nitrogen for 20 min. The NMR data were taken after cooling the sample to room temperature subsequent to each increase of 50 °C in the annealing temperature. The spectra of annealed samples were independent of temperature down to 80 K and field in the range 6.3–1.3 T. All spin-lattice relaxation rates ($T_1^{-1} = R_1$) were also found to be field inde pendent in the same range.

III. RESULTS

A. Proton spin-lattice relaxation under conditions of suppressed homonuclear dipolar interactions

Figure 1 shows the T_{1y} decay of the magnetization for a high (29 at.%) proton content sample (100% SiH_4, 2 W, RT, A). The decay is clearly nonexponential. The curves are from models discussed later in the text. Figure 2 shows the temperature dependence of the initial and final slopes of the T_{1y} decay on an Arrhenius plot. The activation energies from Fig. 2 are 276 ± 30 cal mol^{-1} for the initial slope and 214 ± 50 cal mol^{-1} for the final slope.

Since the two-component dipolar-broadened spectra for these films are due to spatially isolated proton dipolar reservoirs,[7] the nonexponentiality in Fig. 1 may be due to the two proton environments having different relaxation times (T_{1y}). In order to test this hypothesis, the Fourier-transformed, free-induction-decay spectra of the sample shown in Fig. 1 was compared to the spectra after "spin-locking" the magnetization under T_{1y} conditions for a time equal to three times the initial T_{1y} of Fig. 1. Figure 3 shows the "spin-

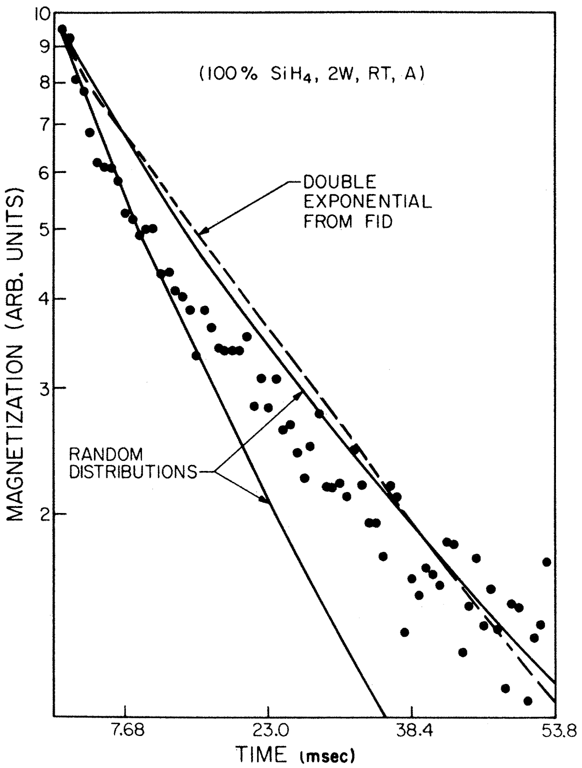

FIG. 1. Decay of magnetization while suppressing homonuclear dipolar interactions (T_{1y}) for an a-Si : H sample prepared under the conditions shown. The dotted line is the sum of two exponentials weighted by the hydrogen content in the broad and narrow components of the Fourier-transformed free-induction decay. The two solid lines are calculations based on the random distribution of relaxation centers discussed in the text.

locked’ spectrum (A), the normal spectrum (B), and the difference. A fit of both spectra to the sum of a Gaussian (broad component) and a Lorentzian (narrow component) yields spectrum A as 87.6% broad (12.4% narrow) and spectrum B as 80.0% broad (20.0% narrow). Figure 3 thus shows the narrow component, on the average, has a shorter T_{1y} than the broad component.

B. The effect of deuteration on T_1 and T_{1y}

Table I shows the deposition conditions for four samples which may be considered in pairs. Each of the two pairs has deposition conditions that differ only by their dilutant gas. In one case the gas is hydrogen and, in the other, deuterium. Neglecting isotopic effects in the plasma chemistry during film growth, Table I shows that between 20–40% of the hydrogen has been replaced by deuterium when films are deposited in deuterium. Included in Table I are the spin-lattice relaxation rates (T_1^{-1}), as well as the initial and final rates

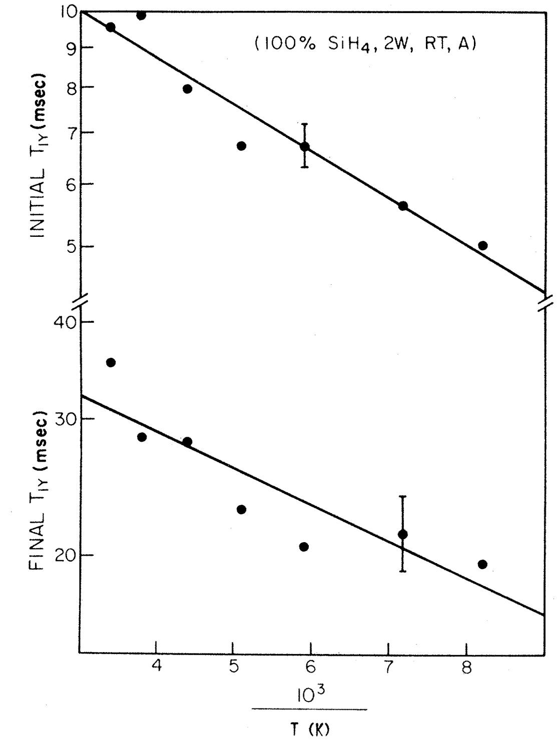

FIG. 2. Initial and final slopes from T_{1y} data as a function of temperature. The activation energy from the initial decay (top) is 276 ±30 cal mol^{-1}; the final decay (bottom) is 214 ±50 cal mol.

(T_{1y}^{-1}) when homonuclear dipole-dipole interactions are suppressed. Figures 4 and 5 show T_{1y} data for the four samples described in Table I. These data clearly show T_1^{-1} to be proportional to hydrogen content and T_{1y}^{-1} almost insensitive to a factor-of-3 change in hydrogen content.

C. The effect of annealing on T_1

Two samples, (5% SiH_4-Ar, 18 W, RT, C) and (5% SiH_4-Ar, 18 W, RT, A), were annealed and monitored for changes in hydrogen content and T_1. Figure 6 shows the changes in hydrogen content for the two samples normalized to unity (as deposited, the samples have different hydrogen contents[7]). The apparent increase in the hydrogen content in the anode sample is not due to adsorption of atmospheric gases, as the sample was kept under dry nitrogen. Also, extended evacuation did not affect the NMR line shape or hydrogen content. Figure 7 shows a comparison of the spin density determined by electron spin resonance (ESR) (from Ref. 9) and T_1 for the two annealed samples. Although the anode T_1 data follow the spin density, the sensitivity is quite low; a change of two orders of magnitude in spin density produces less than a factor-of-2 change in T_1. The cathode sample T_1

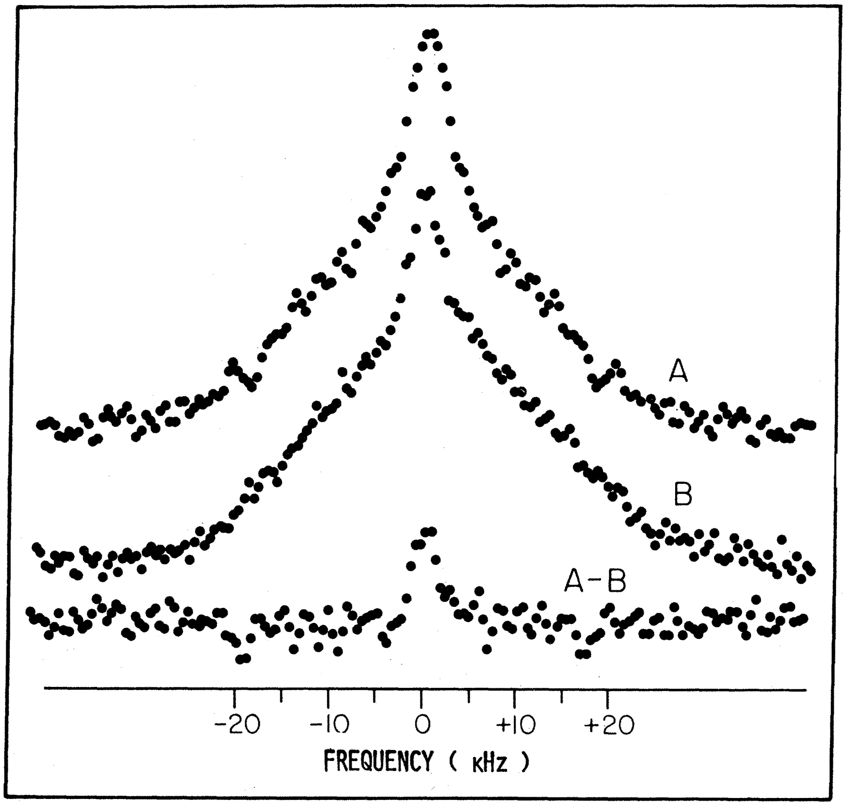

FIG. 3. Fourier-transformed free-induction decays for the (100% SiH_4, 2 W, RT, *A*) sample. Spectrum *A* has been "spin-locked" under the conditions of the T_{1y} experiment (suppressed homonuclear spin diffusion) for a period of three times the initial decay time (Fig. 1) and less than the final decay time. Spectrum *B* resulted from a single 90° pulse. At the bottom is the difference of *A* and *B*.

TABLE I. Deuterium dilution effects on NMR parameters.

Sample[a]	H (at. %)[b]	R_1[c] (sec^{-1})	R_{1y} (initial)[c] (sec^{-1})	R_{1y} (final)[c] (sec^{-1})	$\frac{H_2 \text{ (at. \%)}}{D_2 \text{ (at. \%)}}$	$\frac{R_1 H_2}{R_1 D_2}$	$\frac{R_{1y}^{\text{initial}} H_2}{R_{1y}^{\text{initial}} D_2}$	$\frac{R_{1y}^{\text{final}} H_2}{R_{1y}^{\text{final}} D_2}$
5% SiH_4/H_2, 19 W, RT, *A*	25.7	0.63 ±0.01	31.6 ±0.1	10.6 ±0.1	2.5	2.4	1.0	1.3
5% SiH_4/D_2, 18 W, RT, *A*	10.1	0.2 ±0.01	32.5 ±0.1	8.1 ±0.01				
5% SiH_4/H_2, 18 W, RT, *C*	31.1	0.67 ±0.1	29.3 ±0.1	6.1 ±0.1	4.4	4.0	1.2	2.2
5% SiH_4/D_2, 18 W, RT, *C*	7.0	0.17 ±0.01	23.5 ±0.1	2.8 ±0.1				

[a] Notation for sample-preparation conditions is explained in the text.

[b] Determined from integrated proton spin density and sample weight.

[c] $R_1 = 1/T_1$; $R_{1y} = 1/T_{1y}$.

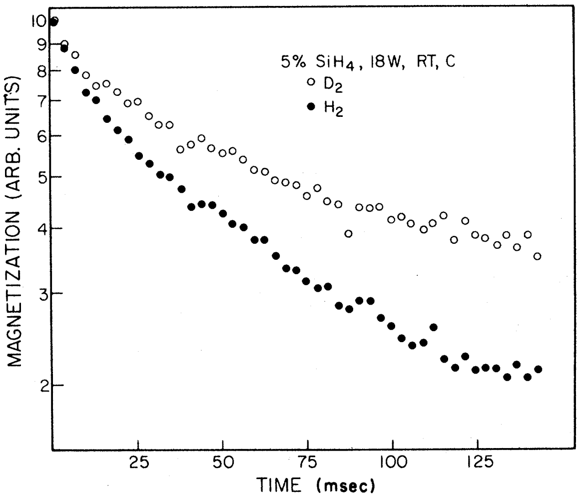

FIG. 4. Comparison of T_{1y} decays for the samples deposited in deuterium and hydrogen on the hot rf electrode (cathode).

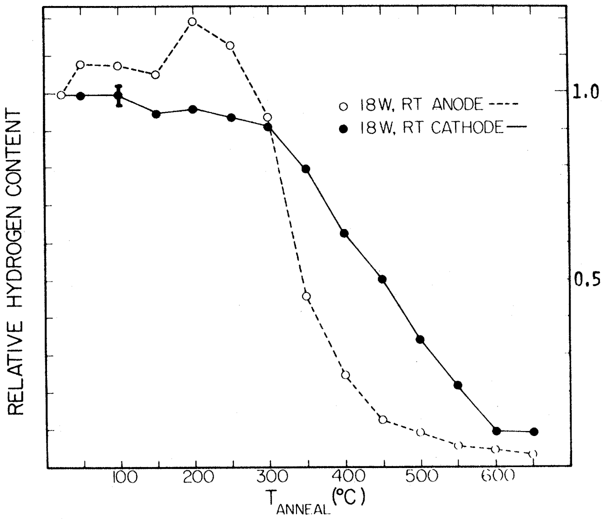

FIG. 6. Hydrogen content as a function of annealing temperature for two films prepared from 5% SiH_4 in argon.

data have no straightforward relation to the spin density.

IV. DISCUSSION

A. T_{1y} and disorder modes

A recent study[10] of NMR spin-lattice relaxation rates (T_1^{-1}) in a-Si:H films has shown that there is a maximum near 40 K and that the temperature and frequency dependence of T_1 are well described by a model which assumes that a small fraction of the hydrogen atoms are coupled to disorder modes. One consequence of this model is that the observed relaxation rate is the result of two processes: (i) spin relaxation for nuclei at the disorder mode, and (ii) spin diffusion that occurs between neighboring spins. The model is analogous to relaxation in solids due to dilute paramagnetic centers[17] where spin diffusion carries to remote nuclear spins the information on the lattice temperature dispensed by electron spins. In these films, however, the role of the paramagnetic center is played by hydrogen nuclei experiencing rapidly fluctuating magnetic fields due to hopping over the potential barrier separating two equilibrium positions of a disorder mode.

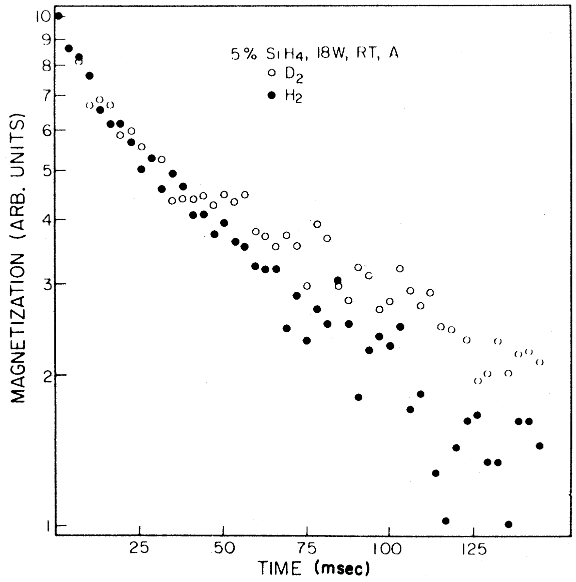

FIG. 5. Comparison of T_{1y} decays for samples deposited in deuterium and hydrogen on the grounded electrode (anode).

Recent developments[14,18] in multiple-pulse schemes for high-resolution NMR in solids[19,20] have furnished a means to measure proton spin-lattice relaxation times while suppressing homonuclear dipolar interactions with an eight-pulse cycle.[15] This experiment allows for a determination of spin-lattice relaxation in a-Si:H in the absence of spin diffusion.[21,22] According to the above model, such a measurement is expected to be analogous to spin-lattice relaxation of isotopically dilute spins, such as ^{13}C, by randomly distributed paramagnetic centers.[23] Nuclei close to the disorder mode are expected to relax quickly, while nuclei far away should relax more slowly since,[14] in the fast motion limit,

$$\frac{1}{T_{1y}} = \tfrac{2}{3}\Delta m_2 \tau \propto \frac{A}{r^6}\tau , \qquad (1)$$

where τ is the correlation time of the hydrogen nuclei in the disorder mode, Δm_2 the fluctuating

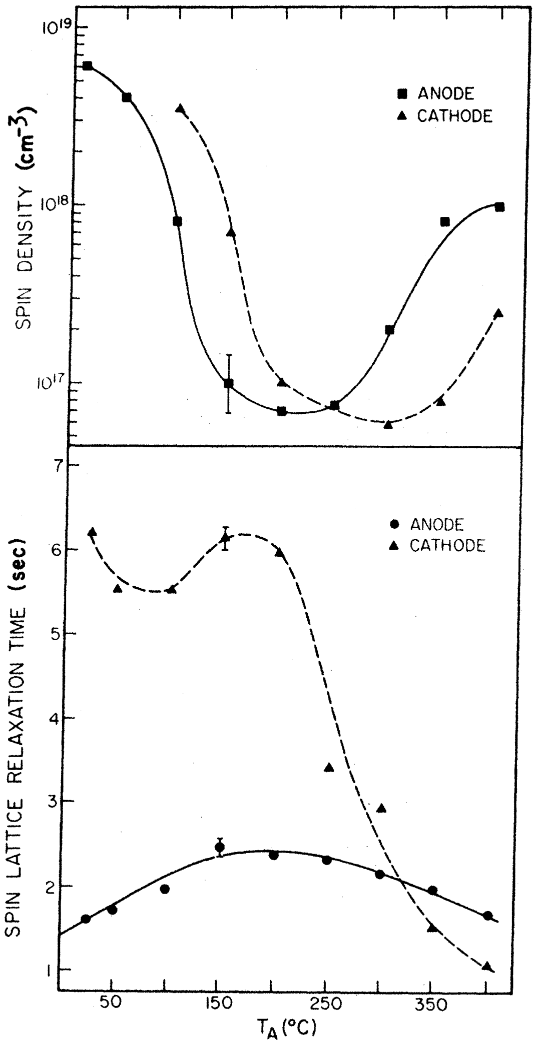

FIG. 7. Proton spin-lattice relaxation times (T_1, bottom) for the samples shown in Fig. 6 as a function of annealing temperature. For comparison the ESR spin densities for similarly prepared samples (from Ref. 9) are shown at top.

field due to hydrogen in the mode, and r the distance between the disorder mode and the nucleus being relaxed. In a disordered material such as a-Si:H one might expect a random distribution of the disorder modes so than an expression $N(T_{1y})$ may be written based on Eq. (1) which gives the number of nuclei at a given T_{1y}. The magnetization then behaves as[23]

$$m(t)=\int_0^{T_{1y}^{\max}} N(T_{1y})e^{-t/T_{1y}}dT_{1y}\,. \qquad (2)$$

One thus expects a nonexponential decay of the magnetization.

The data in Fig. 1 show that the decay of the magnetization in the T_{1y} experiment is indeed nonexponential. This is consistent with the disorder-mode model. Also, if we assume τ is due to a simple activated process,

$$\tau=\tau_0 e^{\Delta E/kT}\,, \qquad (3)$$

then a plot of $\ln(T_{1y})$ versus $1/T$ yields ΔE. Figure 2 shows the initial and final T_{1y} times to be consistent with Arrhenius behavior. The calculated ΔE of ~250 cal mol^{-1} is slightly larger than that observed in the T_1 measurements (Ref. 10), ~160 cal mol^{-1}. However, such differences are to be expected as sample-preparation conditions differ between the two experiments.

Further insight on the distribution of disorder modes can be derived from these T_{1y} data according to explicit models for $N(T_{1y})$ given in Eq. (2). Since the hydrogen atoms are in two spatially isolated regions of the film with differing spin densities,[7] a particularly simple model may be the sum of two exponentials in which the weighting of the two terms is given by the proportion of hydrogen in the high- and low-density hydrogen domains and the time constants of the terms are given by the initial and final time constants in the T_{1y} decay shown in Fig. 1 (Ref. 32). This calculation yields the dotted line in Fig. 1; clearly, this is a poor match to experimentally observed decay.

A second model for Eq. (2) is one in which the disorder modes are distributed at random throughout the film. Using Eq. (1), $N(T_{1y})$ may be calculated readily, and $T_{1y}^{\max}$ is determined by a fit to the data. The solid lines in Fig. 1 show this calculation for two different values of $T_{1y}^{\max}$, one to best fit the initial decay and the second to best fit the final decay. Neither is a good fit to the full range of experimental data. Clearly, these models for $N(T_{1y})$ in Eq. (2) are insufficient to explain the data. The most appropriate model may be one which involves modifying Eq. (2) to include two terms that differ only by $N(T_{1y})$. More precisely, two random distributions may be considered with the average density of disorder modes in each distribution differing. An interesting question would then be whether or not the two different domains of disorder-mode density correspond to the two different domains of hydrogen density. The results for an experiment designed to test this hypothesis are shown in Fig. 3. By "T_{1y} spin-locking" the magnetization for a period of three times the initial time constant, we remove from the spectrum those components which decay quickly. If one domain of hydrogen has a higher density of disorder modes than the other, we would expect the line shape due to that domain to be depleted relative to the other line shape by spin-locking. Figure 3 shows that the spin-locked spectrum for the sample shown in Fig. 1 is depleted in the narrow component relative to the normal spectrum. We conclude that the density of defect sites is higher in the less clustered hydrogen domain. The line-shape analyses show that the average density of hydrogen-containing disorder modes is a factor of 1.3 higher in the less clustered hydrogen domain.

B. Deuterium dilution

At room temperature (far above the T_1 minimum) the spin-lattice rate $T_1^{-1}(R_1)$ is expected to be governed by spin diffusion.[10,17] The spin-diffusion constant[24] is proportional to density of proton spins. Thus, deuterium dilution of the hydrogen in a-Si:H would proportionately affect T_1^{-1}. Assuming there are no isotopic effects during film growth, Table I shows hydrogen content and R_1 to be directly proportional upon dilution of hydrogen with deuterium. These data confirm the importance of spin diffusion on the observed R_1 data away from the T_1 minimum. The T_{1y} data, however, are very insensitive to deuterium dilution. Since the T_{1y} data are not affected by spin diffusion, any changes or lack of changes would be due to the distribution of hydrogen about the disorder modes [Eq. (1) and $N(T_{1y})$ in Eq. (2)]. If the disorder modes contained only one hydrogen nucleus, then the density of hydrogen-containing disorder modes (and T_{1y}) would be proportional to the hydrogen content. Inspection of Figs. 4 and 5 as well as Table I shows that T_{1y} is not proportional to hydrogen content. We conclude that each disorder mode must contain more than one hydrogen atom. This rules out the possibility that the disorder mode corresponds to a single monohydride (SiH) species rapidly exchanging with an unpaired spin or dangling-bond defect.

C. Annealing behavior of T_1

The role the dangling-bond defects play in spin-lattice relaxation is not straightforward. This is made clear by inspection of Figs. 6 and 7, which show changes in hydrogen content and T_1 as the films are annealed. Previous studies[8,9,25-27] have shown changes in ESR spin densities, vibrational spectra, and proton NMR line shapes upon annealing. The following is a summary of the data from this work:

(i) Despite the fact that the unpaired spin density is the same as deposited for the anode and cathode films, the T_1's differ by a factor of ~4.

(ii) In the annealing range of 20–200 °C, where unpaired spin densities rapidly decrease, the T_1's for the anode sample increase whereas the cathode film T_1's are roughly constant.

(iii) In the range 200–400 °C where unpaired spin densities increase, both anode and cathode T_1's decrease.

(iv) The observed hydrogen content (as determined by NMR intensities) for the anode sample increases in the range 20–200. When allowed to self-anneal at room temperature for several months, the hydrogen content in this sample stabilizes at ~32%, with slight changes in hydrogen clustering.[28] The hydrogen content in the cathode sample changes in a straightforward fashion described previously.[8]

(v) A previous ESR study[29] has shown that ele tron spin line shapes in anode samples motionally narrowed for $N_s > 10^{19}$ cm^{-3}. The electron spin T_1's are, however, still too long[10,17] to explain these proton T_1 data. Thus, we may rule out effects on proton relaxation due to paramagnetic centers.[17]

(vi) Anode films have shorter T_{1y} values than cathode films deposited under the same conditions, and the anode film T_{1y} values are less sensitive to deuteration than cathode film values.

In view of these observations, we propose that a fraction of the unpaired spins is associated with the disorder modes and, in anode films, hydrogen nuclei are clustered nearer to the disorder modes than in cathode films. This would explain points (i), (iv), and (vi). The close proximity of hydrogen nuclei to the unpaired spins in the anode film would broaden those proton spectra to the point that they would appear to lack any contribution to the observed line shape. Hence, an artifically low hydrogen content would be observed. The increased local density of disorder modes would yield shorter T_1's, T_{1y}'s, and less sensitivity to deuterium dilution as observed in this study. Observation (ii) may be explained by noting that in the range 20–200 the total number of unpaired spins, and hence the number of disorder modes that contain some fraction of these spins, is rapidly decreasing. The anode film T_1's, sensitive to the density of disorder modes, would then increase. The cathode film T_1's are insensitive to the decrease in unpaired spins (hence, density of disorder modes) due to the proton clustering away from disorder-mode sites (as deposited, longer T_1's, T_{1y}'s, and more sensitive to deuteration). The interpretations for observation (iii) are unclear although we have shown previously,[8] in cathode films at annealing temperatures $T_A > 200$ °C, that hydrogen diffuses internally and thus allows hydrogen nuclei to diffuse closer, on the average, to disorder modes. We conclude that it is likely that the hydrogen-containing disorder modes responsible for proton spin-lattice relaxation may be associated with a small fraction of the unpaired electron spins. The relationship of the different microstructures seen in anode and cathode films *vis-à-vis* the distribution of hydrogen about the disorder modes should be considered.

V. CONCLUSIONS

By using deuterium dilution and spin-lattice relaxation times, while suppressing homonuclear dipolar interactions, we have obtained evidence supporting a hydrogen-containing disorder-mode model of proton spin-lattice relaxation in plasma-deposited amorphous silicon-hydrogen films. A small fraction of hydrogen nuclei at these disorder modes have very short relaxation times and act as "sinks" for proton spin-lattice relaxation. The temperature dependence of these data agree with the T_1 measurement of Carlos and Taylor.[10] Deuterium dilution experiments have shown that: (i) spin diffusion between neighboring nuclei dominates T_1 relaxation at temperatures far above the T_1 minimum, and (ii) each disorder mode contains more than one hydrogen atom. Effects on the proton line shape due to spin-locking, while suppressing homonuclear dipolar interactions, show that the density of disorder modes is ~30% higher in the less clustered hydrogen domain. Finally, we have argued from the T_1 behavior upon annealing that, in addition to hydrogen nuclei, the disorder modes may contain a small fraction of the unpaired spin(s). The relationship between film microstructure, the distribution of the disorder modes, and the optical properties of these films warrants further investigation.

ACKNOWLEDGMENTS

The authors wish to thank R. A. Lujan for his expert assistance with sample preparations. The authors are grateful to Dr. S. I. Chan and Dr. T. M. Duncan for their help in preparing this manuscript, and to Dr. A. J. Vega for helpful discussions. This work was supported by the National Science Foundation under Grant No. DMR-77-21394.

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*Current address: IBM T. J. Watson Research Center, Yorktown Heights, N. Y. 10598.

†Deceased.

[1] H. Fritzsche, C. C. Tsai, and P. Persans, Solid State Technol., 1978, (1), p. 55.

[2] M. H. Brodsky and D. Kaplan, J. Non-Cryst. Solids, 32, 431 (1979).

[3] W. Paul, A. J. Lewis, G. A. N. Connell, and T. D. Moustakas, Solid State Commun. 20, 969 (1976); G. A. N. Connell and J. R. Pawlik, Phys. Rev. B 13, 787 (1976).

[4] W. F. Spear, P. G. Le Comber, S. Kinmond, and M. H. Brodsky, Appl. Phys. Lett. 28, 105 (1976).

[5] D. E. Carlson and C. R. Wronski, Appl. Phys. Lett. 28, 671 (1976).

[6] D. E. Carlson, IEEE Trans. Electron Dev. ED-24, 449 (1977).

[7] J. A. Reimer, R. W. Vaughan, and J. C. Knights, Phys. Rev. Lett. 44, 193 (1980).

[8] J. A. Reimer, R. W. Vaughan, and J. C. Knights, Solid State Commun., in press.

[9] D. K. Biegelsen, R. A. Street, C. C. Tsai, and J. C. Knights, Phys. Rev. B 20, 4839 (1979).

[10] W. E. Carlso and P. C. Taylor, Phys. Rev. Lett. 45, 358 (1980).

[11] J. A. Reimer, R. W. Vaughan, J. L. Knights, and R. A. Lujan, Appl. Phys. Lett., in press.

[12] R. W. Vaughan, D. D. Elleman, L. M. Stacey, W.-K. Rhim, and J. W. Lee, Rev. Sci. Instrum. 43, 1356 (1972).

[13] T. C. Farrar and E. D. Becker, *Pulse and Fourier Transform NMR* (Academic, New York, 1971).

[14] A. J. Vega and R. W. Vaughan, J. Chem. Phys. 68, 1958 (1978).

[15] W.-K. Rhim, D. D. Elleman, and R. W. Vaughan, J. Chem. Phys. 59, 3740 (1973).

[16] R. A. Street, J. C. Knights, and D. K. Biegelsen, Phys. Rev. B 18, 1880 (1978).

[17] A. Abragam, *The Principles of Nuclear Magnetism* (Clarendon, Oxford, 1961).

[18] A. J. Vega, A. D. English, and W. Mahler, J. Magn. Reson. 37, 107 (1980).

[19] U. Haeberlen, Adv. Magn. Reson. Suppl. 1 (1976).

[20] M. Mehring, Nucl. Magn. Reson. Basic Principles Prog. 11 (1976).

[21] D. Tse and S. R. Hartmann, Phys. Rev. Lett. 21, 511 (1968).

[22] J. I. Kaplan, Phys. Rev. B 3, 604 (1971).

[23] T. M. Duncan, J. T. Yates, Jr., and R. W. Vaughan, J. Chem. Phys., 73, 975 (1980).

[24] Reference 17 shows $D \sim Wa^2$, where W is the probability of mutual spin flips between neighbors $(\sim\sqrt{M_2}/30)$ separated by distance a. Light deuterium dilution is expected to change the second moment $(M_2 \propto 1/r^6)$, but the average value of a should remain roughly constant.

[25] C. C. Tsai and H. Fritzsche, Solar Energy Mat. 1, 29 (1979).

[26] J. C. Knights and R. A. Lujan, Appl. Phys. Lett. 35, 244 (1979).

[27] A. J. Leadbetter, A. A. M. Rashid, R. M. Richardson, A. F. Wright, and J. C. Knights, Solid State Commun. 33, 973 (1980).

[28] J. A. Reimer, R. W. Vaughan, and J. C. Knights (unpublished).

[29] R. S. Title, M. H. Brodsky, and J. J. Cuomo, in *Proceedings of the 7th International Conference on Amorphous and Liquid Semiconductors*, edited by E. W. Spear (University of Edinburgh, Edinburgh, 1977), p. 424.

[30] A. J. Vega and A. D. English, Macromolecules 13, 1635 (1980).

[31] M. Goldman, *Spin Temperature and Nuclear Magnetic Resonance in Solids* (Oxford University Press, London, 1970).

[32] $T_{1y})_{\text{initial}} \sim T_{1y})_a[(p_1 + p_b)/p_a]$, where p_a and p_b are the populations of the narrow and broad components, respectively.