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PASSIVATION

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Si(Li) X-RAY DETECTORS WITH AMORPHOUS SILICON PASSIVATION

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Abstract

Lithium-drifted silicon [Si(Li)] detectors with thin lithium n^+ contacts and amorphous silicon (a -Si) junction passivation are described. These detectors (7 mm thick, 9 cm² area) are intended for use in a six-element detector array which is designed to measure trace amounts of plutonium in soil samples. Results are given showing a spectral resolution of ~ 400 eV (FWHM) for the 17.8 keV $N_{K\alpha}$ x-rays entering through either the lithium n^+ contact or the gold barrier contact on these detectors. Measurements on the effects of the fractional H_2 concentration on the electrical behavior of the a -Si/Si interface are reported. The increase with time in the lithium window thickness when the detectors are stored at room temperature is discussed.

Introduction

Interest in improving the monitoring capabilities for trace radioactive nuclides in the environment has prompted our development of specialized x-ray spectrometers. These systems are characterized by having a close sample-detector geometry and a lithium-drifted silicon [Si(Li)] detector with thin entrance contacts on both sides as shown in Fig. 1. The performance of a single detector spectrometer has been reported earlier¹; in this paper we describe modifications to our Si(Li) detector fabrication process to facilitate the assembly of a spectrometer having six detectors, each 9 cm² in area and 7 mm thick.

The fabrication of thin lithium window Si(Li) detectors for charged particle telescopes has been described previously² and this technology was used to fabricate detectors for the initial single-detector spectrometers used for measuring trace amounts of plutonium in soil samples. However, the six-detector spectrometer presents more difficult problems in detector uniformity and stability. We addressed the uniformity problem by employing crystals expected to be defect free³ and the stability question by examining new surface passivation and storage techniques.

The successful application of hydrogenated amorphous germanium (a -Ge:H) to the passivation of high-purity germanium detectors⁴ suggested that amorphous silicon (a -Si:H) might be equally successful in passivating Si(Li) detector surfaces. The use of a -Si:H has been reported⁵ in the passivation of p-n junctions used at room temperature. We report here on the a -Si passivation of Si(Li) detectors, employing various concentrations of hydrogen in the a -Si deposition.

Detector Design

Figure 2 illustrates the salient features of our standard and thin-window Si(Li) detectors. The thin-window detector is fabricated by removing the thick lithium contact which is present on the standard device and rediffusing to produce the thin lithium contact as shown. The details of this process have been discussed previously⁶.

While Fig. 2 indicates that the detector surfaces are protected by KMER[®], passivation of these surfaces presents special problems. Silicon dioxide, which is used almost exclusively for passivation in silicon device technology, cannot be used on lithium-drifted

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Fig. 1. Simplified cross section of the Si(Li) soil analyzing detector system. For clarity, the preamplifier electronics and the isolation of the detector from the cold finger have been omitted.

Fig. 2. Standard and thin window Si(Li) detectors. Shown are the cross sections of a detector with the Si(Li) compensated regions, Li-n⁺ and Au-blocking contacts and KMER[®] passivation.

detectors due to the high temperatures required for its formation. Consequently, Si(Li) detector fabrication has depended on various organic coatings to passivate these surfaces. Our experience with these coatings has been somewhat mixed, with problems of long-term stability, poor adherence, and added 1/f noise being encountered. These problems are compounded when the detector must operate at liquid

[®]Reference to a company or product name does not imply approval or recommendation of the product by the University of California or the U. S. Department of Energy to the exclusion of others that may be suitable.

nitrogen (LN) temperature. Since the coefficient of expansion of the organic coatings does not match that of silicon, cracking and crazing of the organic coatings frequently occur when the detectors are cycled to LN. To circumvent these problems, we have occasionally left the detector surfaces bare, depending solely on the high vacuum in the cryostat to maintain the detector surfaces. However, during the development of the Si-detector spectrometer it became apparent that neither bare silicon surfaces nor organic coatings would be adequate for the detectors in this system.

A second problem encountered with the soil analyzing detectors was the desire to maintain the Li^+ contact as thin as possible. However, when the detectors are stored at room temperature the lithium contact can grow initially at a rate of $4 - 10 \mu\text{m}$ per year, which, if the detectors are stored at room temperature for a few months can lead to a 15 to 20% change in the lithium contact thickness. The growth of these windows is discussed in the following section.

Finally, the close proximity in the soil analyzing cryostat of the preamplifier electronics to the detector bias voltage necessitated the reconfiguring of these detectors as shown in Fig. 3. In our x-ray systems, negative voltage is applied to the detector as indicated in the figure. Consequently, negative bias potential appears on the periphery of the detector as shown by a heavy line in the figure. Any change in the distance between the input signal lead and the detector periphery leads to a charge, ΔQ , being induced into the signal lead, i.e.:

$$\Delta Q = C_S \Delta V_b + \Delta C_S V_b = \Delta C_S V_b$$

where V_b is the bias voltage and C_S is the stray capacitance as indicated in the figure. By cutting down the rim of the detector as shown, we found we could greatly reduce the microphonics in the soil monitor system. The details of this and other aspects of the electronic and mechanical design will be discussed in a subsequent paper.

Fig. 3. Modified thin window detector geometry. The detector geometry modification reduced the stray capacitance, C_S , between the detector rim and the preamplifier input signal lead.

Li⁺ Contact

Figure 4 illustrates the growth of the Li^+ contact thickness with time for two Si(Li) detectors. The contact thicknesses were measured using a ^{228}Th alpha

source to determine the energy loss in the lithium n^+ layer.⁷

The theoretical growth of the lithium contact with time is a mathematically difficult problem to solve. For our purposes, the results obtained by considering this as a two-step diffusion problem⁸ will suffice. The results of this analysis show that the lithium distribution at the contact can be represented by the complementary error function. Consequently, the lithium contact thickness, x_j , during the second "diffusion" can be expressed as:

$$x_j = x_{j0} + bt^{1/2} \quad (1)$$

where x_{j0} is the contact thickness after the first diffusion, t is the second diffusion time, and $b = 2\sqrt{D}$ where D is the lithium diffusion coefficient at the second diffusion temperature (i.e. room temperature). In Fig. 4 the data points have been fitted with $b = 9 \mu\text{m} \cdot \text{year}^{-1/2}$.

The diffusion coefficient of lithium in silicon is⁹:

$$D = 2.3 \times 10^{-3} \exp(-0.645 e/kT) \text{ cm}^2 \text{ sec}^{-1} \quad (2)$$

where $e = 1.6 \times 10^{-19} \text{ C}$, $k = 1.38 \times 10^{-23} \text{ J/K}$ and $T = 293 \text{ K}$ at room temperature. Therefore:

$D = 1.69 \times 10^{-14} \text{ cm}^2 \text{ sec}^{-1} = 59.5 \mu\text{m}^2 \text{ year}^{-1}$ at room temperature.

Hence $b = 2\sqrt{D} = 15.4 \mu\text{m} \cdot \text{year}^{-1/2}$. The agreement between the experimental and theoretical values for b is good considering that the lithium concentration was assumed to remain constant in the derivation of Eq. 1. Lithium in the contact region is supersaturated at room temperature and precipitation will occur, thereby reducing the concentration gradient which is driving the diffusion process. The fact that the experimental value for b is less than the theoretical value indicates precipitation is occurring in these detectors. Further, since the lithium precipitation rate is dependent on the presence of oxygen and point defects, it should be expected that the growth rate of the lithium contact will be variable from crystal to crystal. Crystals with more precipitation sites will exhibit a lower lithium contact growth rate than those with fewer sites. This has been indicated in some detectors which were fabricated on crystals which have defects present³. On these detectors a value of $b = 3.6 \pm 0.5 \mu\text{m} \cdot \text{year}^{-1/2}$ was obtained.

Since the growth of the lithium n^+ contact is dependent on the diffusion coefficient, storage of these devices in LN will diminish the growth rate. Consequently the detectors intended for the soil analyzing x-ray system have been stored in LN. This storage technique placed additional emphasis on the surface passivation to permit cycling between room temperature and LN.

Detector Passivation

As noted earlier, hydrogenated amorphous silicon and germanium have been employed to passivate semiconductor device surfaces. In examining the performance of these coatings, we employed a high resolution x-ray system described previously¹⁰ and small capacitance ($\sim 1 \text{ pF}$) Si(Li) detectors. For the different $\alpha\text{-Si:H}$ coated detectors reported here there was no appreciable difference in the system resolution at 17 μs amplifier peaking time. All the detectors measured produced resolutions of the order of 125 - 135 eV FWHM at LN temperature. There was, however, a marked

Fig. 4. Growth of the thin Li n⁺ contact when detectors are stored at room temperature. The data points have been fitted with the parabolic equation given in the figure.

difference in the leakage current as a function of temperature as shown in Fig. 5. Since the leakage current, I_L , for a reverse biased junction is given by¹¹:

$$I_L = \frac{qn_i}{\tau_0} V \quad (3)$$

where V is the detector volume, τ_0 = the minority carrier lifetime, and n_i = the intrinsic carrier concentration. Now $n_i \propto \exp(-E_g/2kT)$ so that:

$$I_L \propto \exp(-E_g/2kT) \quad (4)$$

where E_g is the semiconductor bandgap energy. As shown in Fig. 5, the slope, obtained using Eq. 4, of the leakage current for the bare surface detector is 1.08 eV in good agreement with the known bandgap energy for silicon of 1.1 eV. Measurement of the slope of the leakage current on the a-Si:H coated detectors yielded activation energies, E_a , as shown in the figure. The table below lists the activation energies we obtained on several small area Si(Li) detectors for different hydrogen concentrations in the argon atmosphere present during the rf sputtering of a-Si onto the Si(Li) detector surfaces.

H ₂ (%)	0.5	7.0	17.5
E_a (eV)	0.62±0.36	0.87±0.17	1.01±0.11

The leakage current of our "best" chemically prepared bare Si(Li) detector surface has an activation energy $E_a = 1.1$ eV = E_g . Our results indicate that the 17.5% a-Si:H coating produces leakage currents with an activation energy comparable to that of our best chemically obtained surfaces. However, the actual leakage current is two orders of magnitude greater at high temperatures. Further examination of these detectors employing Deep Level Transient Spectroscopy (DLTS) using a light to activate the surface states⁴ yielded the results shown in Fig. 6. The response labelled 17.5% H₂ is identical to the response of bare surfaces.

While we have observed the effects of the hydrogen concentration on the leakage current and on the DLTS spectra, neither we nor others¹² have yet described a model which would explain our results and additional work is required to understand the role of a-Si:H on the silicon surface states. Furthermore, it is not

Fig. 5. Detector leakage current, I_L , as a function of $1/T$ (K^{-1}) for detectors with a bare surface and 0.5%, 7.0% and 17.5% a-Si:H coatings. Also shown are the estimated activation energies, E_a , for these currents.

clear from our measurements which coating is to be preferred since it appears that over a selected temperature range, lower concentrations of hydrogen may produce lower leakage currents than higher hydrogen concentrations.

All of the a-Si coatings do appear to produce very stable surfaces. The small capacitance test devices were left for six months at room temperature and in room air. When retested, their performance was essentially unchanged. In addition, repeated cyclings from room to cryogenic temperatures have not produced any change in the test detector characteristics.

While our rf sputtering apparatus has been described previously⁴, it should be noted that rf sputtering is a complicated process with susceptibility to variabilities in process control. For example, the presence of air or water along with the argon atmosphere will result in SiO_x being deposited rather than a-Si. In addition, heat associated with the rf sputtering can produce temperatures sufficiently high to cause the redistribution of the lithium within the Si(Li) detector. Consequently, while we have successfully employed a-Si:H to passivate Si(Li) detector surfaces, we have encountered problems on occasion with the quality of the coating and with decompensation of the lithium. While decompensation of the lithium is fairly easily determined by measuring the change in the voltage required to deplete the detector, the quality of the a-Si:H coating is, at present, more difficult to assess. With germanium detectors, the device geometry facilitates scanning down the sides to assess the

Fig. 6. Deep Level Transient⁴ response of Si(Li) detectors with 0.5%, 7.0% and 17.5% a-Si:H coatings. The 17.5% response is identical with that of our bare surface Si(Li) detectors. The vertical axis is the DLTS output in arbitrary units.

a-Ge:H coating effectiveness⁴. To accomplish similar scans on Si(Li) detectors would require a departure from the silicon detector geometry which is designed to pinch off the detector surface channel¹³. Our few attempts to make Si(Li) detectors without the "pinch-off" design have not been successful, and therefore we presently rely on detector noise, spectral performance and when possible, leakage current-temperature response and DLTS spectra to assess the a-Si:H coating.

Detector Performance

The results of the electronic noise measurements (FWHM) for several soil analyzing detectors with 7.0% a-Si:H coating are shown in Fig. 7. This figure illustrates both the detector performance and the difficulties in assessing whether the coating is contributing to the noise performance. At very long amplifier time constants (> 17 μ sec) where surface 1/f noise produced by the coating can predominate, the system is very susceptible to microphonics and other electronic interference. Consequently, in making the measurements on these detectors, great care was taken to ensure that extraneous signals were not affecting our results. All final measurements on the detectors were conducted with the cryostat system in an rf shielded room. Even so, the differences in the noise performance between various detectors at long amplifier time constants, as evident in Fig. 7, are such that it is difficult to be certain that they are due to the a-Si:H coating or to other factors. The leakage current of the detectors shown in Fig. 7 was approximately 0.1 pA with 800 volts applied bias. In the figure the expected noise due to a 1.0 pA leakage current is indicated. Since the measured leakage currents are an order of magnitude lower than the 1.0 pA curve in the figure, the noise due to the detector leakage current is negligible in the measurements shown.

Spectra of Kp L x-rays from ^{241}Am incident on the lithium and the gold barrier contacts of a Si(Li) soil analyzing detector are shown in Figs. 8a and 8b respectively. Comparison of the two spectra shows that the

Fig. 7. Electronic noise measurements (FWHM) on four 7 mm thick 9 cm² area Si(Li) detectors as a function of amplifier peaking time. The applied bias was ~ 800 V in each case. The expected noise due to $I_L = 1$ pA is also shown.

peak-to-valley ratio for the L_{α} x-rays is slightly poorer for incidence on the lithium contact than on the gold contact (6.5 versus 7.6). This particular detector has been used for a year in a single detector system measuring trace amounts of plutonium in soil samples.¹⁴ The system stability has been excellent indicating that the a-Si:H is effectively passivating the detector junction surface.

Conclusions

Our results have demonstrated that a-Si:H can be successfully employed to produce Si(Li) detectors with very stable low leakage current surfaces at cryogenic temperatures. With the 7 mm thick 9 cm² area soil monitor detectors, the noise at the amplifier peaking time constant of 70 μ sec is being dominated by system microphonics or extraneous signals and not by detector leakage current.

The role of the hydrogen concentration in the a-Si:H on the leakage current is not well understood. While it would appear that a-Si sputtered in an argon atmosphere containing 17.5% hydrogen produces silicon surfaces equivalent to the best that we can obtain chemically, lower concentrations of hydrogen may

result in lower leakage currents over selected low temperature ranges.

Finally, the growth rate of the lithium contacts has been identified as being dependent on crystal defects. Crystals with low defect densities can be expected to have a greater contact growth rate than those with high defect densities.

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Fig. 8. Spectral response of a thin lithium contact Si(Li) detector for ^{237}Th L x-rays from an ^{241}Am source incident on the lithium contact (A) and on the gold contact (B).

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