Submicron entrance windows for an ultrasoft x-ray camera

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During the development of an imaging proportional counter suited for observing celestial objects in the XUV waveband (10–250 Å), we investigated the feasibility of producing a reliable plastic entrance window having a thickness $\leq 0.3\,\mu$ and good uniformity over an area of several tens of square centimeters. After evaluating several candidate materials and production techniques, we selected film casting of pure Lexan as an appropriate technique for providing strong 65 mm diameter, 0.3-μ thick windows having low gas diffusion leakage ($< 3.10^{-4}$ bar cm³/s cm²) and adequate opaqueness for short wavelength UV radiation. A prototype XUV camera equipped with such a window was used to make a picture of a multipinhole mask irradiated by the emission line spectrum of ionized He (243, 256, and 304 Å). This gave very satisfactory performance, as is shown at the end of this paper.

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INTRODUCTION

During the past few years the Cosmic Ray Working Group at Leiden, The Netherlands, has been involved in the development of an imaging proportional counter having high spatial resolution (FWHM $\sim 100\,\mu$ at 1 keV), optimized for use as an ultrasoft x-ray camera in the focal plane of a grazing incidence telescope. Such an instrument, because of its low intrinsic noise and high quantum efficiency, provides the optimum means for the study of extended low brightness celestial sources in the wavelength range of 10–250 Å (XUV range). Examples of such sources are very evolved supernova remnants, emission features from hot (10^6 K) interstellar plasma, and the alleged hot galactic halo.

Until now, the operation of imaging proportional counters as focal plane cameras has been mainly restricted to wavelengths shorter than ~80 Å, which is determined by the thickness of the counter entrance window. In order to shift this long wavelength cut off to beyond 200 Å, an ultrathin plastic window, with a thickness of 0.3 μ or less, had to be developed which meets certain requirements concerning effective area, uniformity, mechanical strength, diffusion leak, and wavelength pass band. The initial development of such a window, which would eventually qualify for space application, was undertaken in cooperation with the Röntgenbuizenlaboratorium of Philips Nederland B.V. and results of this development are reported here.

The window for such a camera should have an effective area of ≈ 35 cm² and good uniformity in thickness (variation < 10%). Figure 1 shows the transmission efficiency as a function of photon wavelength for 0.5- and 1-μ thick polypropylene (state of the art in rocket and satellite experiments, respectively) and of 0.3- and 0.15-μ thick polycarbonate foils of Lexan, and demonstrates the required increase in long wavelength response.

Furthermore, the following requirements were imposed on the window properties: (A) the gas diffusion leak, due to permeability, must be $< 3.10^{-4}$ cm³ bar/s cm² at a differential pressure across the window of 0.2 bar. This requirement arises, among others, from the limited gas storage possibilities in space experiments. In addition, the supported window should
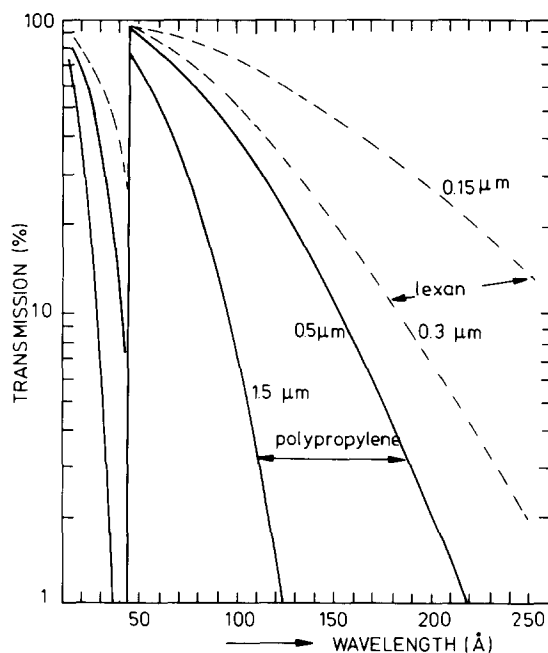

FIG. 1. XUV transmission as a function of wavelength for polypropylene and Lexan foils of various thickness.

withstand pressure cycling between 0 and 1 bar (several hundred times), and temperature cycling between $0°$ and $40°$ centrigrade without noticeable increase of the diffusion leakage. (B) One side of the window must be conductive to allow for the application of a well-defined electric field inside the counter drift region. The thickness of a conductive coating should however, not impair the XUV-transmission properties. (C) The window should efficiently absorb UV photons with wavelengths shorter than $\simeq 2500$ Å. The quantum energy of these photons (>5 eV) exceeds the photoelectric work function of conductors, thereby releasing single electrons from the coating into the counter gas. The UV component of stellar XUV sources can therefore introduce a serious source of background if this precaution is not implemented, since the tail of these single electron distributions extends into the XUV range.

I. EVALUATION OF PRODUCTION TECHNIQUES

Several methods have been employed in the past for the production of thin plastic foils, both for the use as beam filters and as entrance windows for gas counters. For the latter application, $0.5~\mu$ polypropylene has been successfully used in rocket borne soft x-ray experiments[1-3] and can therefore be regarded as the present state of the art for space application.

To investigate the feasibility of making a much thinner window, we first explored several different production techniques to get an empirical feel for their specific limitations and critical areas. We shall now proceed with a brief account of our experience, after which a more detailed description of the adopted production technique and performance measurements will be given.

Biaxial stretching of polypropylene $(C_3H_6)_n$ foils has proven to be an excellent method for the production of thin counter windows.[1] For areas of several tens of square centimeters, however, the limit of this technique seems to be a thickness of about $0.3~\mu$. This limit can be reached by using a rather sophisticated and thermally well-tuned set-up as described by Barres and Blake.[4] Since polypropylene transmits UV radiation down to 1600 Å, an additional coating for UV absorption is necessary anyhow, and consequently the window would effectively come close to a thickness of $0.5~\mu$, which has too much XUV absorption. This production technique has therefore been discarded.

Evaporation techniques have been used to produce ultrathin (1000 Å) metallic foils.[5] However, these can only be used as beam filters and are much too weak to withstand substantial differential pressure.

Vacuum vapor-phase polymerization has been successful for the production of highly uniform Parylene N pellicles. Parylene N is a linear polymer $(C_8H_8)_n$, based on poly-p-xylylene.[6,7] Since these pellicles are commercially available at a thickness of 1000 Å up to 10 cm diameter, we conducted leak tests on a few small sample pellicles (supplied by Union Carbide) mounted on a fine support mesh (see Sec. 3). They all showed, however,

a leak rate far in excess of our requirement. The main difficulty encountered with this technique was releasing the parylene from the supporting glass subtrate. Since the prohibitive leak rate is most likely associated with this separation process, we did investigate this problem in some depth. With the aid of an existing facility and available expertise at the Philips Research Laboratory, we produced several batches of Parylene N pellicles (method according to Spivack[7]). On a very smooth glass substrate (irregularities < 50 nm) a thin interface layer was deposited before the polymerization process of Parylene was performed, in order to facilitate a fast and nondestructive separation process. After several tests with different interface layers, we finally chose a double layer composed of a low molecular release agent, covered by high molecular Poly-Vinyl Alcohol. The release agent provides a fast release process whilst the PVA layer prevents degradation of the polymerization process by the high vapor pressure of the release agent. Separation can be easily accomplished by dipping in a 1 : 1 solution of isobutanol and water at $60°$C. In this way several highly uniform $0.15-0.3~\mu$ Parylene N pellicles were produced. However, subsequent leak tests showed that a majority of the samples violated our requirement by an order of magnitude at $0.15~\mu$. In addition, Parylene N transmits UV in the $1400-1800$ Å band[8] and therefore requires coating with an UV absorber. We therefore abandoned production of Parylene N in favor of Lexan (see further on).

A third method involves casting of the plastic film. This method was introduced by Henke,[9] who produced thin Formvar [trademark of Monsanto, mainly $(C_5H_8O_2)_n$] film by casting a Formvar solution (solvent 1.2 dichloroethane) on a water surface. Formvar layers as thin as $0.02~\mu$ can be formed in this way during evaporation of the solvent. Williamson and Maxon[10] have used this method to produce large counter windows (up to 1800 cm²) of $1~\mu$ thick by laminating several layers, which eliminates the effect of small pinholes. In order to reduce the window transmission for UV radiation below 2500 Å, they added the polycarbonate Lexan [trademark of Cadillac Plastic and Chemical Company $(C_{16}H_{14}O_3)_n$] disolved in CADCO-SC-125 (also available from Cadillac Plastic). Lexan is an excellent absorber for UV radiation below 2400 Å (see Fig. 2). Pure Lexan films can also be made in this way and have been used to coat polypropylene windows to reduce their UV transmission.[11] This film casting method is advantageous in two respects. First of all, no supporting substrate is necessary, which eliminates the problem of film release from this substrate. Secondly, the film is built up from several layers which greatly reduces the influence of small pinholes on the leak rate. We tried this method on a batch of candidate materials, i.e., Formvar, Lexan, Bioden (acetyl cellulose), and Collodion (cellulose nitrate), to evaluate which, if any, would have superior mechanical strength and leak rate. Small samples (ϕ 14 mm) with a typical thickness of $0.2~\mu$, comprising 5 layers, were successfully produced for all of these materials. Lexan 101 dis-

solved in dichloromethane with a softener consistently showed the lowest leak rate. Since Lexan is also favored for its low UV transmission, we subsequently concentrated on the production of large (ϕ 65 mm) pure Lexan windows. This did include the investigation of the criticality of a number of process parameters for the quality of the end product.

II. LEXAN WINDOW PRODUCTION TECHNIQUE

Figure 3 shows a picture of the production tooling for Lexan film casting. The device is composed of a water reservoir into which a spreader, containing the Lexan solution, can be lowered. This should be done in such a way that water surface tension pushes the solution upwards along the spreader surface until eventually the solution distributes uniformly over the intersecting line between the spreader and the water surface. This process can be best studied by coloring the solvent with, for example, Rhodamin B. The spreader (width 12 cm) is attached to a carriage which can move along a rail driven by a small servomotor. In this way the spreader is drawn slowly along the entire length of the water tank, leaving a Lexan film floating on the water surface. The Lexan film is formed within a few seconds after spreading on the surface. The quality of the produced film is critically dependent on many system parameters.

First of all, the set up has to be contained in a dust-free environment (less than 50 particles per cubic meter). Distilled water at about 20 °C (which is frequently renewed and led through a dust filter) should be used to avoid the presence of small dust particles on the water surface. Investigation of the formation process of the film on the water surface showed that the best results are obtained by using a brass hydrophobic spreader,
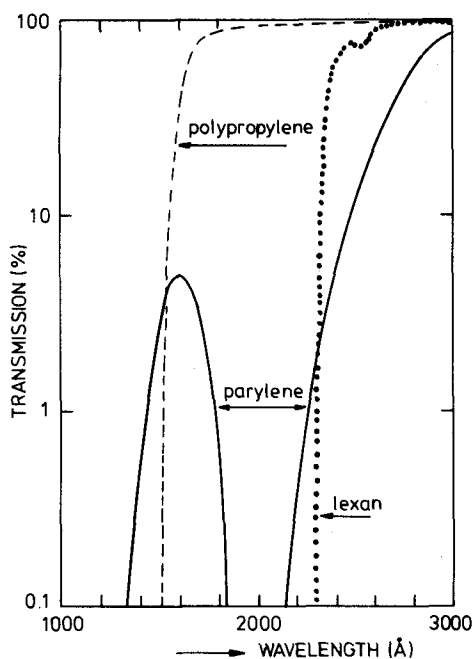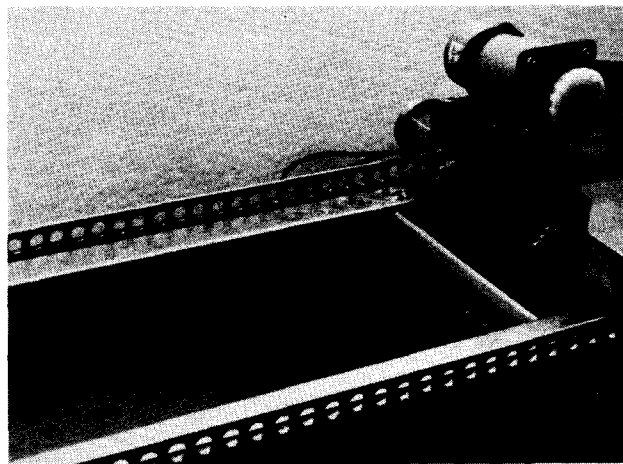

FIG. 3. Production tooling for Lexan film casting. The polymerized Lexan film, floating on the water surface, is just discernible.

which should dip well below the surface. Both the Lexan concentration in the solution and the drawing speed determine the thickness of the film. Analysis of different batches of commercially available CADCO-SC-125 solvent showed slightly different constituents and a high degree of dust pollution. We therefore decided to produce a clean Lexan solution composed of dichloromethane with 0.01% dioctylphtalate as a softener and 2% Lexan 101 powder (by weight). For a lower concentration of Lexan, the removal of a Lexan film from the water surface is much more difficult. A 2% solution in conjunction with a drawing speed of 1 cm/s gave a typical thickness of 700 Å. It is important that all possible sources of vibration or turbulence near the water surface during the time of film casting should be eliminated to avoid nonuniformities in the film thickness. This means adequate decoupling of small vibrations of the carriage motor from the water tank and a shutdown of air circulation in the dust-free bench during the casting process.

After the film is formed on the water surface, a stainless steel mask with an open aperture of 65 mm is lowered onto the film, next dipped into the water starting from one side, and then raised from the tank. The free-standing Lexan film adheres to this mask and can be dried. Repetition of this process between 2 and 5 times yields the desired thickness range of 0.15 μ. It should be noted that, once a basic Lexan film of, say, one or two 700 Å layers is produced, lower concentrations of Lexan can be used to adhere thinner layers to this film, allowing for a trim to the desired thickness. Each individual layer is, before stacking, subjected to microscopic examination for the detection of large pinholes.

It is evident that a free-standing window of this size is capable of withstanding little differential pressure. Assuming a supporting mesh with circular holes, the burst pressure P_B of a film with thickness t relates to the hole diameter D by the relation

$$P_B \simeq \frac{4t}{D} \sigma,$$

FIG. 2. UV transmission of 0.3 μ Paralyne N (cross sections according to Stern and Paresce,[8] Polypropylene according to de Korte,[18] and Lexan (Williamson and Maxon[10] and Yamashita[19]).

with σ the tensile strength of the plastic film. For the plastics under consideration $\sigma \approx 5.10^7$ N/m^2. Imposing $P_B > 1$ bar for a window of 0.15 μm thick, requires a mesh support with a pitch $< 100~\mu$. We selected for this purpose a nickel mesh which is fabricated by means of an electroplating technique which was specifically developed by Philips Nederland B.V. for the production of storage grids in oscilloscope tubes. The mesh we employed had a pitch of 50 μ and a transmission factor of 0.55. This fine mesh was in turn supported by a coarse mesh of etched stainless steel having a pitch of 1 mm and a transmission factor of 0.85. These meshes were contained in an aluminum support assembly which possessed 1-mm thick support bars on a 2-mm pitch to avoid excessive bending of the window-mesh combination over the 65 mm aperture. During the mounting of the window on the support assembly it turned out to be difficult to avoid edge effects which could damage the window.

Application of a conductive coating of 70 Å thick aluminum was successfully accomplished by vapor phase deposition at 5.10^{-8} bar. This thickness does not impair the transmission for XUV radiation, except for a small region around 170 Å (Al L-edge), where the absorption reaches a maximum of 25%. The uniformity of this coating was very good, its thickness being assessed by transmission measurements ($T_{\mathrm{opt}} \approx 20\%$). The conductivity of the coating was $\approx 30~\Omega$/cm, which is more than adequate. It would be desirable to use coatings with a higher photoelectric work potential than Al to decrease the UV sensitivity. We have not yet pursued this any further.

III. QUALIFICATION PROCEDURE

Qualification of the produced windows with respect to strength and gas diffusion leak was accomplished in the following way. The leak rate of the supported window was first evaluated at a differential pressure of 0.2 bar. Subsequently the window was subjected to pressure cycling (typically 100$\times$) applying progressively higher differential pressures from 0.2 to 1 bar, if intermediate checks do not show deterioration. Afterwards the leak rate was assessed at 0.2 bar and had to be less than 3.10^{-4} cm^3 bar/s cm^2 for qualification. The leakage of a window is due to both small pinholes and the intrinsic diffusion leak. The lowest measured leak rate is an upper limit for the intrinsic diffusion leakage and turned out to be less than 5.10^{-5} cm^3 bar/s cm^2 for two samples with a diameter of 65 mm and a thickness of 0.3 μ. Qualification tests on a batch of 50 0.3 μ thick 65 mm diam. window samples showed a yield of 25%. We did not succeed in producing qualified windows of this size with a thickness of 0.15 μ; this, however, may still have been due to imperfections in the mounting procedure on the support structure and therefore 0.3 μ should not be regarded as a fundamental limit.

In addition the XUV transmission of the window has to be assessed. Given the chemical composition, the thickness can serve as a reasonable indicator. The thickness and uniformity of the window material can be judged in the most straightforward way by the color of the light reflected from the window for white light at normal incidence.[12] This method has an absolute accuracy of $\pm 0.02~\mu$ in the thickness range from 0.1 to 0.3 μ. Thickness nonuniformities can be clearly distinguished down to 0.01 μ. With the aid of dual beam UV interference, the color-thickness relation can be calibrated ("color-thickness"). Table I gives the color-thickness for a few Formvar and Lexan samples for which the XUV transmission has also been measured. Inspection of the color pattern was used to ensure that the desired uniformity over the window aperture was indeed obtained.

A direct measure of the XUV transmission was obtained with the help of a Penning discharge source. Filled with helium gas, this source emits the Lyman series of ionized helium at 243, 256, and 304 Å. A transmission grating provided the separation of the different lines. The results of these measurements are also given in Table I, together with the XUV transmission values expected from the measured "color-thickness" and the absorption cross sections of the constituent atoms.[13] Although the transmission predicted from the color-thickness and the measured XUV transmission are not inconsistent, it should be noted that all the measured values are consistently lower than the predicted transmission. We want to emphasize in this context that the transmission properties of plastic windows between 100 and 1000 Å can only be derived approximately from the weighted sum of the cross sections of their con-

TABLE I. Color thickness and XUV transmission of Lexan and Formvar samples.

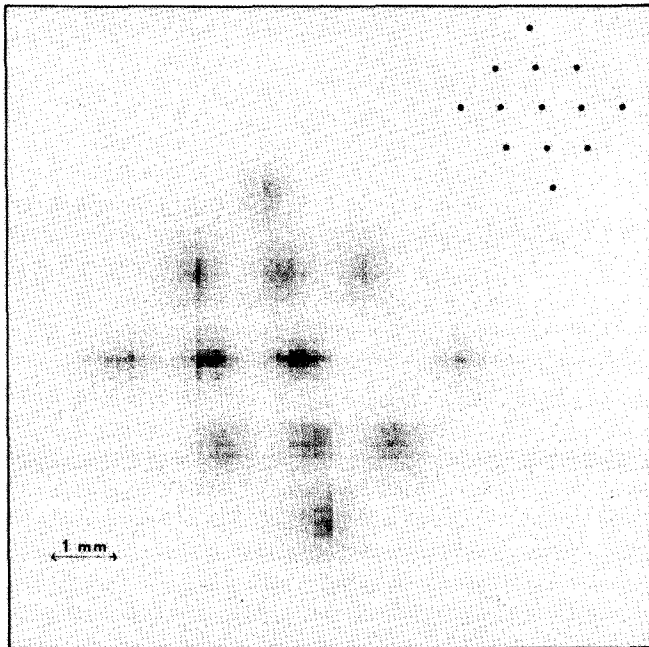| Material	Calibrated color thickness (microns)	Predicted transmission (%)			Measured transmission (%)		
		243 Å	256 Å	304 Å	243 Å	256 Å	304 Å
Lexan	0.11 ± 0.02	25.5^{+7}	23.5^{-7}	15.4^{+6}		16 ± 1	7 ± 1
	0.15 ± 0.02	15.3^{-5}	14.2^{+4}	7.2^{+4}	16 ± 1	9 ± 1	4 ± 1
Formvar	0.11 ± 0.02	22.5^{+7}	20.5^{-6}	12.3^{+6}		14 ± 1	10 ± 1
		(6.2^{-4})	(4.2^{+4})	(2.1^{-2})			
	0.13 ± 0.02	17.1^{+5}	15.1^{-5}	9.3^{+3}	13 ± 1	10 ± 1	4 ± 1
		(4.2^{-2})	(2.1^{+2})	$(1.0.5^{+1})$			

FIG. 4. XUV image at 250 Å of a multipinhole mask taken with a gas camera featuring a 0.3 μ thick Lexan entrance window. The pixel size equals 0.1 mm, the position resolution is about 0.6 mm FWHM. The image spots do not have equal brightness due to nonuniformity in the incident beam and obscuration by the window support structure. The latter causes the dropout of one hole (see insert).

stituent atoms. Above the Carbon K edge (44 Å), the electrons of the L shell play a dominant role in the absorption process. The L shell electrons, in contrast to the K shell electrons, are not uniquely localized at the atoms (valence electrons). Lenselink[14] showed that due to this effect the molecular cross section of Formvar may have to be raised significantly for wavelengths near 300 Å as compared to the weighted sum of the atomic cross sections. In Table I the predicted transmission according to his model is given for Formvar in parentheses. They, in turn, are consistently lower than the measured transmission, indicating that rather large uncertainties still exist in making reliable transmission estimates based on thickness information.

IV. AN XUV GAS CAMERA

A prototype XUV gas camera, based on the detection principle described by Bleeker et al.,[15] has now been fully tested by the Cosmic Ray Working Group at Leiden over the wavelength range 10–300 Å. This camera has an effective entrance diameter of 65 mm diameter and shows excellent linearity and uniformity over the full aperture ($\approx$35 cm^2). The position resolution measured at 1 bar ranges from 80 μ (FWHM) at 10 Å to 600 μ at 250 Å. A detailed description of this instrument and its performance will be given elsewhere[16,17]; however, we show here some first results obtained near 250 Å when the camera was equipped with a qualified 0.3 μ Lexan window.

In front of the camera a multiple pinhole mask (pitch 1.3 mm, hole size 0.2 mm) was mounted which was irradiated (in vacuo) by a parallel beam comprising the emission line spectrum of ionized helium. Independent measurements of the transmission of the window showed that equal intensities of 243/256 and 304 Å radiation were injected into the camera which was operated at 0.4 bar. The pinhole mask pattern and its XUV image, registered by the camera, is shown in Fig. 4.

[1] T. Kato, T. Kohno, F. Nagase, K. Nishimura, Y. Tanaka, and K. Yamashita, Jpn. J. Appl. Phys. 11, 1351 (1972).
[2] P. A. J. de Korte, J. A. M. Bleeker, A. J. M. Deerenberg, Y. Tanaka, and K. Yamashita, Astrophys. J. Lett. 190, L5 (1974).
[3] J. A. M. Bleeker, J. Davelaar, A. J. M. Deerenberg, H. Huizenga, A. C. Brinkman, J. Heise, Y. Tanaka, S. Hayakawa, and K. Yamashita, Astron. Astrophys. 69, 145 (1978).
[4] D. M. Barrus and R. L. Blake, Rev. Sci. Instrum. 48, 116 (1977).
[5] W. R. Hunter, D. W. Angel, and R. Tousey, Appl. Opt. 4, 891 (1965).
[6] W. F. Gorham, J. Polym. Sci. 4, 3027 (1966).
[7] M. A. Spivack, Rev. Sci. Instrum. 41, 1614 (1970).
[8] R. Stern and F. Paresce, J. Opt. Soc. Am. 65, 1515 (1975).
[9] B. L. Henke, Adv. X-Ray Anal. 8, 269 (1964).
[10] F. Williamson and C. W. Maxon, Rev. Sci. Instrum. 46, 50 (1975).
[11] D. G. Fabricant, R. E. Goddard, F. R. Harnden, and P. Gorenstein, Rev. Sci. Instrum. 50, 727 (1979).
[12] M. Francon, Handbuch der Physik, XXIV, 208 (1956).
[13] R. Cruddace, F. Paresce, S. Bowyer, and M. Lampton, Astrophys. J. 187, 497 (1974).
[14] A. J. Lenselink, internal report Cosmic Ray Working Group, Leiden, The Netherlands (1979).
[15] J. A. M. Bleeker, H. Huizenga, A. J. F. den Boggende, and A. C. Brinkman, Proc. IEEE Trans. Nucl. Sci. NS-27, 176 (1980).
[16] H. Huizenga, Ph.D. thesis, University of Amsterdam (1980).
[17] H. Huizenga and J. A. M. Bleeker, Nucl. Instrum. Methods (to be published).
[18] P. A. J. de Korte (private communication).
[19] K. Yamashita (private communication).