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Moderation and diffusion of positrons in tungsten meshes and foils

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The efficiency of tungsten meshes and thin foils for moderation of fast positrons from ^{22}Na has been investigated in transmission geometry and a fair agreement has been found with previous experimental results where directly comparable. For foils, the dependence on material thickness is found to be similar to the prediction of the Vehanen-Mäkinen diffusion model; however, the magnitude is 5–10 times lower. A broad consensus is observed between experiment and the results of a three-dimensional model developed in this work. For a given thickness, meshes are found to be generally better than foils by around a factor of 10 with a maximum efficiency ($\sim 10^{-3}$) comparable to that achieved with thin single crystal foils, in accord with previous measurements and the results of the present model.
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I. INTRODUCTION

Moderators are a crucial component for the production of quasi-monoenergetic positron beams, reducing the energy of β^+ particles emitted from radioactive decay (or pair production) down to a few electron volts (e.g., Schultz and Lynn 1988, Coleman *et al*. 2000). Whilst the highest moderation efficiencies (almost 1%) are given by rare gas solids (RGS) (Mills and Gullikson 1986, Gullikson and Mills 1986, Greaves and Surko 1996), the requirements for their operation are not trivial, the cryogenic conditions being particularly cumbersome in an electrostatic beam (e.g., Massoumi *et al*. 1991). The most commonly used alternatives are W meshes or foils. Although historically around two orders of magnitude less efficient than RGS, W moderators are simpler to handle and remain stable for an extended period of time. However, significantly higher efficiencies ($\epsilon_m \sim 10^{-3}$) have more recently been obtained from stacks of meshes which had been electropolished (Saito *et al*. 2002) or etched (Weng *et al*. 2004). The latter authors found the efficiency to depend on the annealing pre-treatments, the duration of the etching and the number of the folding layers. Stimulated by these observations, and in the course of the development of a new electrostatic beamline at UCL (Kövér *et al*. 2014), we have investigated the variation of ϵ_m for W foils (both single- and poly-crystals) and meshes with material thickness and annealing pre-treatments. A model has also been developed to allow for three-dimensional (3D) diffusion of the thermalized positrons and its results are compared with present and other available measurements.

II. EXPERIMENTAL METHOD

The experimental investigations have been performed using an electrostatic beam (Kövér *et al*. 2014, Williams *et al*. 2010, and Kövér *et al*. 2010). Briefly, fast β^+ particles (N_{β^+}) from ^{22}Na impinge onto the moderator under investigation. The re-emitted slow positrons are then accelerated to 2 keV and focused to a spot size of radius 1 mm on a remoderator where brightness-enhancement (Mills 1980) is performed in a transmission geometry by using a 500 angstrom thick W foil (Jacobsen *et al*. 1990). The remoderated positrons (N_+) are then accelerated to the required energy, transported through zoom lenses, and deflected through 90° by a cylindrical mirror analyser to separate them from fast particles and γ-rays emanating from the source region. A position sensitive detector (PSD) monitors the positrons at the end of the flight path. Secondary electrons released at the remoderator are detected using a channel electron multiplier (mounted off the beam axis), triggering the start of a timing sequence which is terminated by the positron arrival at the PSD.

In order to compare the experimental moderator efficiency with theoretical predictions, as full account as possible must be made of the various factors (such as geometry, source efficiency, etc.) which may affect the measured moderation efficiency. As described in detail by Kövér *et al*. (2014), a coincidence method can be applied to calculate the number of moderated positrons (N_P). The method employs the measured number of remoderated positrons at the end of the beam line (N_+), that of secondary electrons emitted from the remoderator (N_-) and the number of coincidences between detected secondary electrons and positrons (N_c). These can be expressed as

$$N_+ = \varepsilon_+ N_P, \tag{1a}$$

$$N_- = \varepsilon_- N_P, \tag{1b}$$

$$N_c = \varepsilon_c \varepsilon_+ \varepsilon_- N_P, \tag{1c}$$

where ε_+ is the combined efficiency for positron remoderation, transport, and detection; ε_- is for emission, transport, and detection of the secondary electrons, and ε_c is that for the coincidence electronics (~1 in this work). In this way, $N_P = (N_+ N_- / N_c)$ may be determined, and hence, the corresponding moderation efficiency

$$\epsilon_m = N_P / N_{\beta^+}, \tag{2}$$

where N_{β^+} corresponds to the number of β^+ particles hitting the moderator and corresponding to the source activity (in Bq) multiplied by the positron emission branching ratio (90%), an approximate source efficiency of 24% (as per Table I) and fractional solid angle of 40%. During the course of this work, two different sources were used with activities of 6 mCi (Dupont/New England Nuclear, original activity 138.2 mCi) and 18 mCi (iThemba Labs, original activity 50 mCi).

To test the hypothesis that the moderation efficiency could be increased by using meshes with thinner diameter wires, an etching procedure was devised similar to the one used by Weng *et al.* (2004). The mesh (Swallow Metals: 99.99% purity; wire diameter, $d = 25\ \mu$m; transmission coefficient, $t = 81\%$) was cut into 15 mm diameter circular disks and placed into a sodium hypochlorite solution; d was reduced to ~19 μm (t ~ 86% transmission) in approximately 20 min at 75 °C. The reduction in d was calculated by measuring the mass (m), before (b) and after (a) etching using

$$\frac{m_b}{m_a} = \frac{d_b^2}{d_a^2} \tag{3}$$

and t calculated from

$$t = \frac{(L - d)^2}{L^2}, \tag{4}$$

where L is the wire separation in the mesh stated by the manufacturers. Figure 1 illustrates the variation in mesh transmission with the duration of etching and temperature of the solution. As moderators, meshes are usually stacked together to enhance interception of the β^+ particles emitted by the radioactive source (e.g., Zafar *et al.* 1996). Weng *et al.* (2004)

TABLE I. Relevant parameters for the sources used in this work. Their initial activities are shown in brackets.

	DuPont/ NEN (138.2 mCi)	IThemba (52.7 mCi)
Thickness of Ti window (μm)	13	5
Transmission coefficient, T	0.31	0.64
Backing material	Ti	Ta
Backscattering coefficient, B	0.31	0.49
Forward fraction, $F = 0.5(1 + B)$	0.65	0.75
Active diameter (mm)	3	3.7
Self absorption[a] (A)	0.5	0.5
Estimated source efficiency ($T \times F \times A$)	0.1	0.24
Measured source efficiency	0.34[a]	(0.09, 0.31)[b]

[a]This measurement (Massoumi *et al.* 1988) refers to a New England Nuclear (NEN) source of 100 mCi initial activity. The same authors found self-absorption to be negligible for a NEN source of 10 mCi initial activity.
[b]These measurements (Reurings *et al.* 2006) refer to IThemba sources of initial activities 50 mCi and 30 mCi, respectively.

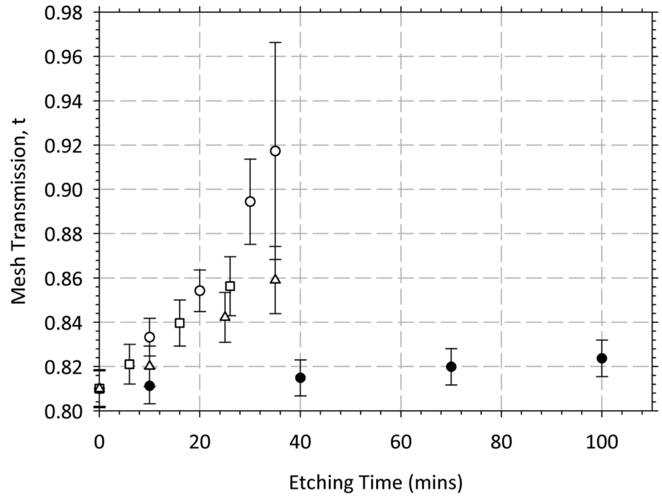

FIG. 1. The increase of the transmission coefficient, t (for meshes with wire diameter 25 μm) as a function of the etching time: (●) at room temperature and (□, Δ, and ○) at 75 °C.

expressed the probability of extracting a slow positron from such an arrangement as

$$P \propto (1 - t) N t^{N-1}, \tag{5}$$

where N is the number of meshes in the stack. Using Eq. (5), the optimum number of meshes for a given t can be estimated, $N_{opt}(t) = 3(0.7)$, 5(0.81), and 7(0.86). As detailed in Sec. III, ε_m was measured for several stacks of etched and unetched meshes, and for W foils of varying thickness and crystallinity. Both mesh and foil moderators were annealed following the procedure described in Zafar *et al.* (1988) During the measurements, a stack of 7 etched meshes (which showed a good stability between tests) was placed in one of the slots in the moderator holder and used as a reference.

III. EXPERIMENTAL RESULTS AND DISCUSSION

The present determinations of the efficiencies for single- and poly-crystal foils as well as etched and unetched meshes are displayed in Figures 2 and 3, respectively.

In Figure 2, the 1.0 μm and 1.8 μm foils were single crystals of orientation (100) (grown at Aarhus University as described in Zafar *et al.* 1988); the 12 μm and 25 μm foils were polycrystalline. A dependence on material thickness similar to that theoretically predicted by Vehanen and Mäkinen (1985) may be discerned. However, the magnitude of the experimental results is generally a factor of (5–10) lower. A reasonable consistency may be observed with previous measurements. However, the most direct comparison is with those of Gramsch *et al.* (1987) and of Zafar *et al.* (1988), since these authors carried out measurements using low-activity open sources (thus obviating the need to correct for self-absorption and window attenuation) and applied similar corrections to ours where applicable. In these works as well as ours, the annealed moderators were exposed to air. As in the work of Lynn *et al.* (1985), Gramsch *et al.* (1987) annealed the moderators *in situ* but further noted that no degradation in efficiency was seen when the film was exposed to air for a few hours. The efficiencies obtained by Lynn *et al.*

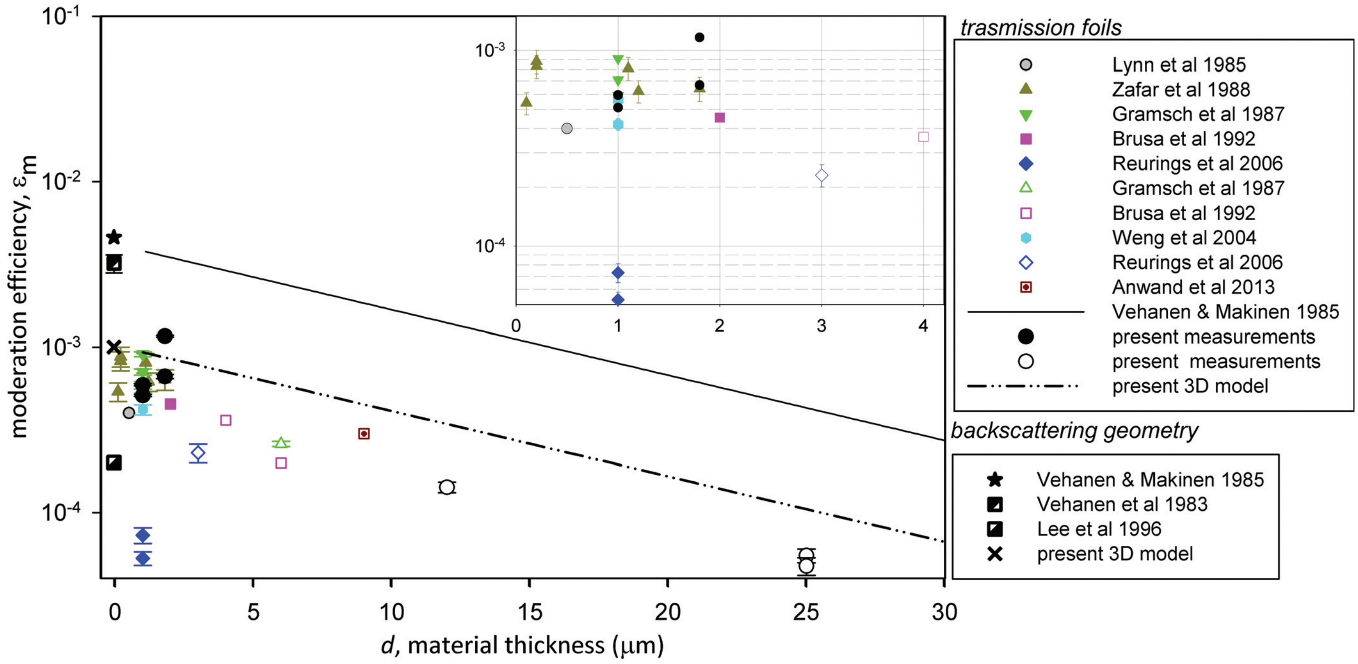

FIG. 2. Moderation efficiency as a function of thickness of W foil transmission moderators: (solid symbols) W (100), (hollow symbols) polycrystalline; (line symbols) theories. The inset zooms in the small *d* values. Also shown (at $d = 0$) are experimental and theoretical results for backscattering moderators.

(1985), Brusa *et al*. (1992), and Weng *et al*. (2004) for the unetched foil are, within the scatter of the data, not inconsistent but, as all other previous experimental determinations, they should be considered underestimates since not all the efficiencies discussed in Section II may have been accounted for in these works.

In comparison with theory (Vehanen and Mäkinen 1985), Lynn *et al*. ascribed the observed discrepancy of their measurements to the low specific activity of their ^{22}Na source whilst Brusa *et al*. to surface conditions. The results of Reurings *et al*. (2006) are around 10 times lower than the other experimental data, the authors mentioning the annealing method as a possible issue.

In combination, we note that the results for the polycrystal foils (hollow circles in Figure 2) extrapolate at $d = 0$ to an efficiency lower than the single crystals by approximately a factor of 2.

As shown in Figure 3, for a given moderator thickness, meshes generally yield efficiencies higher than foils by around a factor of 10, with maximum efficiencies of approximately 10^{-3} comparable to (if not higher than) those achieved with thin single crystal foils, although the planar geometry of the latter makes them better suited for high-brightness applications (e.g., Köver *et al*. 2014). In our work, using the 6 mCi source, 7 etched meshes (t = 0.86, d = 19 μm) were placed in the moderator holder at the same time as 4 unetched meshes (t = 0.81, d = 25 μm). An increase of approximately 25% in the efficiency was observed for the former; however, this is comparable to the increase (23%) in the stack transmission and to fluctuations between samples. Thus, we have no evidence that the moderator efficiency increases with decreasing wire diameters. Also, using the 18 mCi source, a stack of 7 etched W meshes (t = 0.86, d = 19 μm) were found to have an efficiency similar to that measured for a stack of 3 unetched W meshes (t = 0.70, d = 20 μm, as supplied from Swallow Metals) of similar overall transmission (~34%), implying that the efficiency does not depend intrinsically on etching.

IV. DIFFUSION MODEL, RESULTS, AND DISCUSSION

The efficiency $J(d)$ of transmission moderators of thickness d has been evaluated by Vehanen and Mäkinen (1985) by considering a one-dimensional (1D) diffusion equation, obtaining results which, as shown in Figure 2, are a factor (5–10) times larger than experimental data. In order to investigate the reason for this discrepancy, we have at first

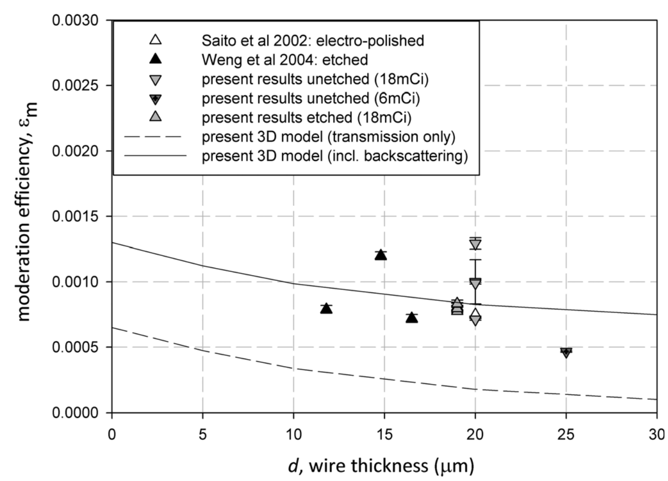

FIG. 3. Moderation efficiency of W meshes as a function of wire thickness. Symbols: (up/down triangles) etched/unetched meshes. Also shown are the results of the 3D model discussed at the end of Section IV.

considered a simplified 1D model for the diffusion process which assumes a simple exponential decay of the form $F(x) = e^{-\frac{1}{L_+}x}$, with L_+ being the diffusion length. In so doing the transmission efficiency is given by

$$J(d) = Y_0 \int_0^d P(z) e^{-\frac{1}{L_+}(d-z)} dz = Y_0 e^{-\frac{1}{L_+}d} \int_0^d \alpha^+ e^{-\left(\alpha^+ - \frac{1}{L_+}\right)z} dz. \tag{6}$$

We have used the same implantation profile (Brandt and Paulin 1977, Mourino *et al*. 1979) as in Vehanen and Mäkinen (1985), $P(z) = \int_0^d \alpha^+ e^{-\alpha^+ z} dz$, where α^+ is the mass absorption coefficient, defined through $\alpha^+/\rho = 2.8 Z^{0.15}/\bar{E}^{1.19} \left(\frac{\text{cm}^2}{\text{g}}\right)$ with $\bar{E}$ being the mean energy of the incoming β^+ particle. We have also used the same values for W as used by Vehanen and Mäkinen (1985), $L_+ = 1350$ Å and $\alpha^+ = 910.53$ cm^{-1}. The branching ratio ($Y_0 = 0.33$) gives the probability that the positron, having diffused to the exit surface, will be emitted as a slow positron (Vehanen *et al*. 1983).

Eq. (3) can be integrated analytically to give

$$J(d) = Y_0 \frac{\alpha^+}{\frac{1}{L_+} - \alpha^+} \left[e^{-\alpha^+ d} - e^{-\frac{1}{L_+}d} \right]. \tag{7}$$

The results of this equation are the same to within (0.01–0.001)% as those of Vehanen and Mäkinen (1985). We can see in Equation (7) that, since $1/L_+ \gg \alpha^+$, $J(d) \sim Y_0 \alpha^+ L_+ [e^{-\alpha^+ d}]$, i.e., the efficiency is approximately linear with respect to the diffusion length. Hence, in order that the results of the 1D models be consistent with the experimental values, the value of L_+ used in Equation (7) would need to be decreased by a factor (5–10), which seems unlikely, even though the value of L_+ will be affected by defects and impurities in the samples.

Both approaches, (i.e., that of Vehanen and Mäkinen (1985) and that given by Equation (7)) are fully one-dimensional in the sense that, with reference to Figure 4, it is assumed that the positrons thermalize at a distance (q) from the surface in a direction ($\hat{z}$) along that of the incoming β^+ particle, itself normal to the surface of the foil. This is consistent with the manner in which the implantation profile has been established experimentally (Brandt and Paulin 1977, Mourino *et al*. 1979). It is then considered that the thermalized positrons diffuse along the same direction, so that the overall distance over which diffusion takes place is just (*d-q*), *d* being the thickness of the foil. It is this simplification which we believe gives rise to the overestimate of the efficiency in the 1D model. By allowing instead the thermalized positrons to diffuse isotropically then, as illustrated in Figure 4, they will follow diffusion paths of lengths $\frac{d-q}{\cos\theta'}$. Therefore, the overall distance over which the moderation takes place may now be longer than d and positrons will be reemitted overall with a lower efficiency. In order to evaluate this new consideration, we have extended the 1D model expressed by Equation (7) to 3D.

With reference to Figure 4, the probability that an incident fast positron stops at a distance q from the entrance surface (i.e., at a point *B)* is given by the implantation profile (Brandt and Paulin 1977, Mourino *et al*. 1979)

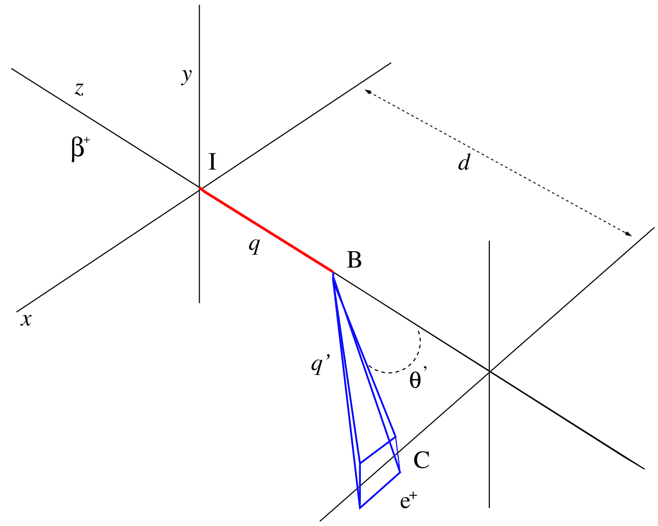

FIG. 4. Geometrical illustration of the 3D description of the moderation process.

$$dP_B = \alpha^+ e^{-\alpha^+ q}. \tag{8}$$

Assuming isotropic diffusion, the probability that the positron will diffuse from B into a small solid angle around the point C on the exit surface is

$$dP_C = dP_B \frac{\sin\theta' d\theta' d\phi'}{4\pi} e^{-\frac{1}{L_+}q'}, \tag{9}$$

where q' and θ' are the length and angle as in Figure 4, related to q by $q' = \frac{d-q}{\cos\theta'}$.

The total probability of a positron reaching the point C on the exit surface is $P_t = P_B P_C$. Hence, given that the efficiency is $J(d) = Y_0 P_t$, we find that, after integration over the azimuthal angle

$$J(d) = \frac{\alpha^+}{2} \int_0^d e^{-\alpha^+ q} \left[\int_0^{\pi/2} e^{-\frac{1}{L_+}\frac{d-q}{\cos\theta'}} \sin\theta' d\theta' \right] q^2 dq. \tag{10}$$

The results computed from Equation (10) for W (using the same parameters as in the 1D calculations) are, as shown in Figure 2, approximately a quarter of those of Vehanen and Mäkinen (1985) and closer to the experimental values, overestimating the efficiency by approximately 30% for single crystals and a factor 2 for polycrystals. Given the higher residual defect concentration expected for the latter, this discrepancy may be genuine and may warrant further investigations. We note also that the changes required in L_+ in order to getter closer correspondence with experiment for these latter foils are now of the order of (40–50)%.

We have extended the model to backscattering geometry and obtained an efficiency J(backscattering) = 0.001, also approximately one quarter of the 1D model prediction. There are only two experimental data (Vehanen *et al*. 1983 and Lee *et al*. 1996) available in this geometry and, as shown in Figure 2, the scatter is so large that it is difficult to draw a firm conclusion from the comparison. Further measurements in this geometry would be useful. We also note that explicitly allowing for different isotopes, in the case of ^{58}Co (used by Vehanen *et al*. 1983), the efficiency predicted by the 3D model increases by 14% with respect to ^{22}Na whilst, in the case of ^{64}Cu (used by Lee *et al*. 1996), it decreases by 15%.

Using the present 3D model, we have further investigated the moderation process for meshes by integrating the contributions from thickness (0 to d) to find the transmission efficiency for a mesh of diameter d. This is shown in Figure 3 where it may be seen to be significantly smaller than experiment. Including the backscattering contribution, i.e., taking the overall moderation efficiency for the meshes as the sum of the integrated transmission and backscattering (multiplied by 0.65 to account for the approximate area presented by the mesh to the incoming β^+ particles), fair accord with experiment is found.

V. CONCLUSIONS

The efficiency of tungsten meshes and thin foils for moderation of fast positrons from ^{22}Na has been investigated in transmission geometry and reasonable agreement has been found with previous experimental results. Whilst the theoretical predictions of a one-dimensional model generally overestimate experiment by up to an order of magnitude, closer correspondence is found between theory and measurements for both foils and meshes across a variety of experiments by extending the diffusion model to three dimensions.

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