Positron dynamics in surface-charged solid argon

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Studies have been made of the reemission of positrons incident at low energies upon solid argon to which electric fields were applied by charging an overlayer of molecular oxygen. An enhancement in positron reemission was observed which reached a maximum for an applied field of around 7 kV mm^{-1}. At this field strength the same yield was observed for implantation energies ranging from 1 to 10 keV, consistent with enhancement due to field-induced positron drift to the exit surface. At higher electric fields, the observed gradual decrease in enhancement was attributed to the heating of the positron energy distribution above the positronium formation threshold. Quantitative agreement with our experimental results has been obtained using a Monte Carlo simulation from which estimates for the positron diffusion length and mobility of $1.7(+2.0,-0.4)$ μm and $4.7(+2.9,-0.4)\times10^{-3}$ $m^2V^{-1}s^{-1}$, respectively, have been derived. This model was also able to successfully reproduce previous results obtained using surface-charged argon β^+ moderators. An abrupt and almost complete reduction in positron reemission was observed for applied surface potentials above a value which showed a weak dependence on film thickness.

I. INTRODUCTION

The study of positron dynamics within solids and at their surface is not only of interest in its own right, but has been the focus of much research over the past 20 years in the development of more efficient β^+ moderators for the production of low-energy positron beams.[1] Much of this research has involved the use of metals in which positron thermalization is generally rapid and precedes diffusion and possible emission into vacuum from a negative work function surface. The rare-gas solids (RGS), despite having positive work functions, were discovered to have good positron reemission properties and to be efficient β^+ moderators.[2,3] Here the dynamics were described in terms of epithermal diffusion and emission of positrons as a result of inefficient cooling below the positronium formation threshold of these wide-band-gap insulators. The highest quoted moderation efficiencies for a flat geometry ($0.30\pm0.02\%$) were obtained using solid neon films.[3] Enhancements in this efficiency have been obtained using different source/moderator geometries with a cup yielding $0.70\pm0.02\%$ (Ref. 3) and a conical design $1.4\pm0.2\%$.[4]

One method of improving moderation efficiencies that does not rely solely on the random diffusion of positrons to an exit surface was originally discussed by Madey[5] and utilizes a bulk electric field in order to drift implanted positrons to the surface of a semiconductor or insulator. The first attempt to construct such a device was made by Lynn and McKee[6] who used a silicon wafer and applied the electric field using a thin gold contact. This and other later attempts have not been successful, probably due to trapping of the positrons at the metal-semiconductor interface.[7,8] More recently Shan *et al.*[9] have reported, from a positron-lifetime-type investigation, that $\sim10\%$ of β^+ implanted into GaAs could be drifted to a metal contact and suggested how it might be possible to design a field-assisted moderator using this technique.

A field-induced enhancement in moderation efficiency was recently observed by Merrison *et al.*[10,11] as a result of applying an electric field to solid Ar and Kr films. The method involved a technique in which the RGS surface was charged by capture of low-energy electrons on adsorbed O_2.[12,13] The work reported here has been carried out as an extension of the latter investigation in order to better understand the positron dynamics in such surface-charged RGS by using beam implantation and observing the subsequent low-energy positron reemission.

II. EXPERIMENTAL SETUP AND PROCEDURE

The study was performed using a magnetically guided positron beam with a ~75 MBq ^{22}Na radioactive source and a W mesh moderator producing a beam of 2×10^4 slow positrons per second. This beam was passed through a trochoidal charged-particle velocity filter at an energy of 100 eV before being accelerated to energies in the range of 1–10 keV. The acceleration was performed by having the entire source end floating at a positive potential equal to the desired beam energy minus 100 V. The beam had an energy spread of ~4 eV full width at half maximum and a diameter of roughly 5 mm as measured using a ceratron lowered into the beam axis immediately in front of the target. In the experiment, im-

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plantation energies of 1, 2, 4, 6, 8, and 10 keV were used.

The target consisted of a copper plate mounted on, but electrically isolated from, the cold finger of an APD Cryogenics model H2 cryopump (Fig. 1). The cold finger was surrounded by a grounded copper radiation shield and cooled to ~ 12 K. A retarding grid inserted to analyze the reemitted beam was placed at a distance of 4.8 mm from the target.

The argon samples were frozen directly onto the copper substrate by letting gas into the chamber at a pressure of 2×10^{-4} Torr for various lengths of time to obtain layers of different thickness. The gas had a stated purity of 99.998% with the major impurities being N_2 (<10 ppmv) and H_2O (<3 ppmv) and was used without further treatment. The base pressure of the vacuum chamber was $\sim 2\times10^{-9}$ Torr. Film thicknesses were calibrated by depositing a relatively thick layer of argon (depositing time ~ 6 h) and measuring the increase in the target voltage required to prevent implantation of 100–450 eV beams of positrons as a result of the argon surface becoming closer to the earthed retarding grid. Through simple capacitance considerations this shift in retarding potential is given by $\Delta V=[d/\varepsilon_r(D-d)]E_{e+}$, where d is the sample thickness, ε_r is the relative dielectric constant for argon, D is the distance between the copper substrate and the earthed retarding grid, and E_{e+} is the positron energy in eV. The measured ΔV was of the order of 5% of E_{e+}. Assuming linearity with depositing time and gas pressure it was then possible to calculate the thickness of the other samples. The thicknesses used in the experiment were thus determined to be 4, 8, 16, 24, and 40 μm with an error of around 20% in all cases, which combines uncertainties from the depth calibration experiment and in the pressure during deposition. This method was also used to estimate the thickness of the oxygen overlayer, which was applied to the sample by letting in this gas at a pressure of 2×10^{-7} Torr, typically for 60 s and thus obtaining a coating of 15 ± 3 Å.

The surface-coated RGS was subsequently irradiated by low-energy electrons from a tungsten filament lowered into the beam axis roughly 2.5 cm in front of the target. The filament was biased at -1 V giving an electron current of 50 nA impinging on the target. During irradiation the target was slowly ramped positively over several minutes, at a rate of ~ 0.5 V s^{-1}, until the desired surface potential was reached, as measured using a 100-eV positron beam by determining the target bias required to prevent implantation. Exposure of the surface to the impinging slow positron beam caused some decay of the surface potential at a rate of roughly 5 V h^{-1}, whereas no decay was observed with the beam off. Recharging was performed simply by repeating the process outlined above.

The low-energy reemitted positron reaction was measured by alternately biasing the target ±45 V with respect to the surface potential while monitoring the positron annihilation rate in the argon using a Ge(Li) γ-ray detector which observed the sample through a lead-slit system. The purpose of the slit arrangement was to reduce the background due to annihilations on the 86% transmission copper retarding grid by preferentially viewing the argon film. Data acquisition times of 300–800 s per point were used to obtain good statistics with increasing accumulation time at the higher energies due to reduced beam intensity.

III. EXPERIMENTAL RESULTS

For a clean uncoated argon sample the reemission was observed to decrease linearly with increasing implantation energy (Fig. 2) in reasonable agreement with previ-

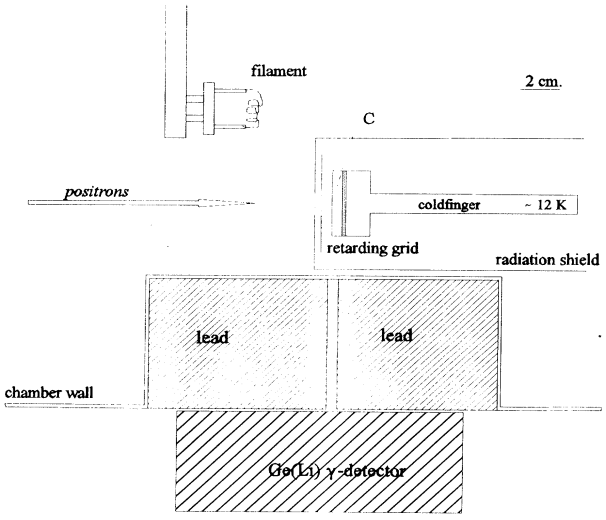

FIG. 1. The experimental setup near the sample. The sample copper backing C is electrically isolated from the cold finger.

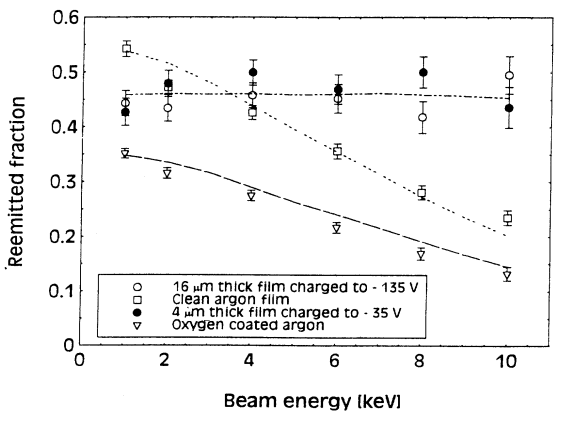

FIG. 2. Reemission yield vs implantation energy for Ar samples subjected to various surface conditions. Note that despite differences in surface potential and thickness the reemission of the two charged samples, which both have electric fields of around 8.5 kV mm^{-1}, is much alike. The lines are plots of the simulation discussed in Sec. IV.

ous studies.[3] The reemitted fraction included only positrons that left the surface of the sample with an energy of less than 45 eV. It did not include backscattered positrons or those emitted as positronium. The backscattered fraction should constitute a little over 10% for solid argon when interpolating the results of Mäkinen *et al.*[14] and Coleman *et al.*[15] to $Z=18$. Figure 2 also shows a decrease in reemission yield after treatment with oxygen, most notably for the lower implantation energies. After irradiating with electrons to produce a negative surface potential a marked increase in the reemitted fraction was observed at higher implantation energies with the yield at 10 keV increasing by a factor of ~ 5. The reemission after charging was observed to be roughly constant over the entire energy range.

As shown in Fig. 3, the positron reemission was observed to increase with surface potential (and thus with bulk electric field) up to a maximum at about 7 kV mm^{-1} for all film thicknesses. For electric fields above this maximum and up to a sudden cutoff, described later, a gradual reduction in the enhancement was observed, which was dependent upon implantation energy, with those implanted at lower energies suffering less reduction in remission yield.

In addition to the effects described above another distinct process was apparent, which is illustrated most clearly in Fig. 3. When a specific value of the surface potential was reached, for a given sample thickness, a rapid decrease in the reemitted fraction was observed for all implantation energies. This cutoff was also observed for positrons implanted at 100 eV. At higher energies the effect is less discernible due to the already low reemission value. The reemitted yield decreased below the level of the uncharged sample resulting in a value close to zero. The surface potential at which this occurred varied weakly with thickness from around -95 V for the thinnest sample (4 μm) to -215 V for the thickest sample (40 μm). For the thicker samples this decrease appeared before the maximum remission enhancement had been achieved.

It is interesting to note that a maximum surface potential was observed above which it was not possible to further charge the sample. This maximum seemed to depend weakly on film thickness, increasing from -170 V for the thinnest films to -220 V for the thickest samples.

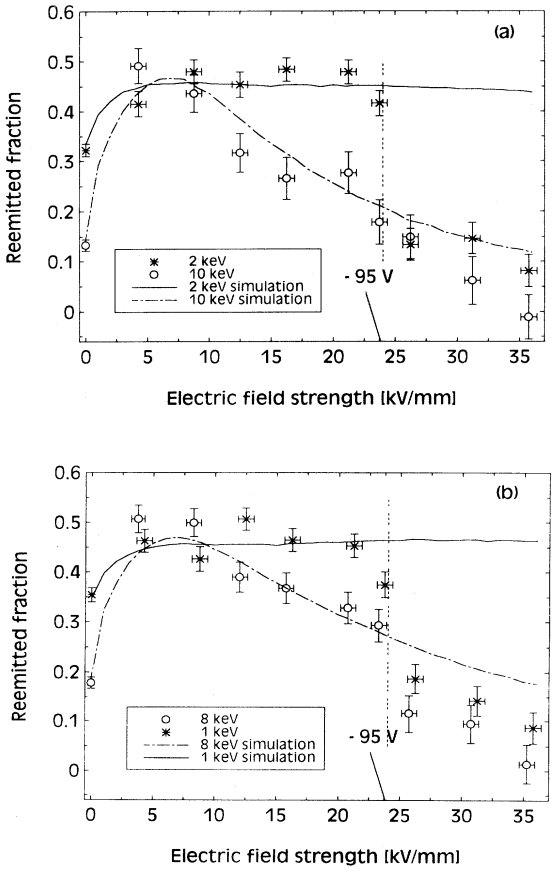

FIG. 3. Reemission yield vs electric field strength for a charged 4-μm-thick O_2-coated Ar sample for implantation energies of (a) 2 and 10 keV and (b) 1 and 8 keV. The lines are plots of the simulation discussed in Sec. IV. The vertical dashed line indicates the approximate onset of the cutoff.

IV. COMPUTER SIMULATION

A computer simulation was carried out in an attempt to model the experimental results. It was not possible to perform a true Monte Carlo calculation as has been done for various metal substrates[16,17] since this requires knowledge of scattering cross sections for all energy loss processes encountered following implantation and these are presently unknown for the RGS. A Makhovian implantation profile with a shape parameter $m=1.9$ was therefore used to describe the positron implantation profile. This was taken to mean slowing to below the bulk inelastic (positronium formation) threshold $E_{\mathrm{Ps}}^{B\mathrm{Ar}}$. The positrons then proceed on a random walk from discrete depths with an energy chosen randomly (below $E_{\mathrm{Ps}}^{B\mathrm{Ar}}$) and in random directions. After traversing a scattering length (l_s) the positrons were isotropically scattered and lost an average energy (dE) due to a phonon interaction. The specific parameters and assumptions used in the simulation are given below.

(1) The Makhovian implantation profile is given by

$$P(z)=\frac{1.9z^{0.9}}{z_0^{1.9}}\exp[-(z/z_0)^{1.9}]\ ,$$

where z_0 is related to the mean implantation depth $\bar{z}$ by $z_0=\bar{z}/\Gamma[(1/1.9)+1]$ where Γ is the gamma function.[18,19] An integration over the Makhovian profile was performed using an ensemble of typically 10^5 positrons. (2) The Ps formation fraction on slowing down was 10%.[20] (3) The bulk positronium threshold for argon, $E_{\mathrm{Ps}}^{B\mathrm{ar}}$, was 10 eV.[2] Positrons gaining energy in excess of this were assumed to form Ps and annihilate in the bulk. The effective Ps thresholds at the surface of O_2-coated Ar, $E_{\mathrm{Ps}}^{O_2}$, and contaminated Ar, $E_{\mathrm{Ps}}^{c\mathrm{Ar}}$, were 5.9($\pm$0.4) and 8.1($\pm$0.4) eV, respectively. These last two values were obtained as best-fit parameters to the experimental data.

(4) The average energy loss per collision, dE, was $1.1(+0.4,-0.5)$ meV. This was used as a fitting parameter to reproduce the experimentally observed reemitted fraction. This value can be compared to the Debye energy of 7.4 meV, corresponding to $\Theta_D = 85$ K, which is a measure of the maximum phonon energy. (5) The scattering length l_s was also a fitting parameter and was taken to be $250(+150, -20)$ Å, in agreement with other work.[21] (6) The value used for the positron work function, ϕ_+, was $+1.5$ eV.[21] Positrons reaching the surface with perpendicular energy below this value were reflected. (7) The positron lifetime in argon was 430 ps.[22] (8) The positron mass was assumed to be m_e, identical to its vacuum value. (9) The effect of the electric field ϵ, which is assumed to act throughout the bulk of the film, was to add a small drift distance in the direction toward the exit surface of the film

$$\Delta z_i = \frac{1}{2}\frac{e\epsilon}{m_e}\tau_i^2$$

and to give the positron an extra velocity in this direction (and thus to increase the energy)

$$\Delta v_i = \frac{1}{2}\frac{e\epsilon}{m_e}\tau_i ,$$

where τ_i is the time between collisions for the ith step and m_e is the electron mass.

It should be noted that this is a rather crude model. It is reasonable to expect that the energy loss per collision is a function of positron energy. The value of dE used in the simulation should therefore be viewed as an average value. Similar arguments hold for the scattering length l_s. Uncertainties quoted on the values of dE, $E_{Ps}^{O_2}$, E_{Ps}^{BAr}, and l_s were estimated by independently varying each parameter while requiring a reasonable fit to the e^+ reemission observed from the Ar samples when clean, O_2 coated, and with applied electric field. In addition to the fraction of reemitted slow positrons the simulation also calculated the fraction of implanted positrons reaching energies below 2 kT and the fraction forming positronium.

Unfortunately the positronic properties of solid oxygen are virtually unknown. There was thus no theoretical foundation to describe the effect of the solid oxygen overlayer. The presence of the oxygen (approximately 10 ML) was taken to lowered the positronium threshold energy, $E_{Ps}^{O_2}$, at the surface. A reasonable fit to the experimental data was found giving a value of $E_{Ps}^{O_2} = 5.9(\pm 0.4)$ eV. The corresponding threshold in the gaseous phase is at 5.3 eV. Given the base pressure ($\sim 10^{-9}$ Torr) and data accumulation time for the "clean" argon results, a surface contamination of at least one monolayer must be anticipated. A modification of the effective Ps threshold at the surface was therefore assumed. A reasonable fit to the data was obtained using a value of $E_{Ps}^{cAr} = 8.1(\pm 0.4)$ eV.

V. DISCUSSION

Generally good agreement was obtained between experiment and the results of the computer simulation, despite the crudeness of the latter (see Figs. 2 and 3). Using the values of dE and the scattering length (l_s) given earlier an estimate of the diffusion length may be obtained: $L = (E_{Ps}^{BAr}/2dE)^{1/2} l_s = 1.7(+2.0,-0.4)$ μm. An estimate may also be made of the positron mobility $\mu = e\tau_{sc}/m$ where τ_{sc} is the average time between collisions. A value for τ_{sc} can be obtained by calculating the mean velocity during thermalization $v_{th} = (E_{Ps}/2m)^{1/2}$, this yielded $\tau_{sc} = l_s/v_{th} = 2.7(+1.6,-0.2)\times 10^{-14}$ s and thus a mobility of $\mu = 4.7(+2.9,-0.4)\times 10^{-3}$ $m^2 V^{-1} s^{-1}$. It should be noted that the value for dE [$1.1(+0.4,-0.5)$ meV] is not in agreement with that obtained previously of $6(\pm 2)$ meV (Ref. 2) and therefore the derived value for the diffusion length is also different. The value for the scattering length [$l_s = 250(+150,-20)$ Å] is in agreement with a previous determination (230 ± 90 Å).[21]

The model suggested that the lower positron reemission yield after coating the Ar film with molecular oxygen (Fig. 2) was the result of positrons with energies larger than $E_{Ps}^{O_2}$ forming Ps at the surface, similar to the effect of the contaminants on the clean argon film. Shallowly implanted positrons (lower incident energy) generally have higher energies on returning to the surface and thus a larger fraction are subsequently lost due to lowering of the Ps formation threshold by the O_2 on the surface.

The observed increase in reemission following charging of the oxygen overlayer and the successful modeling of this is consistent with an efficient field-induced drift of positrons to the surface. After applying an electric field of 7–8 kV mm^{-1} (by surface charging) roughly constant positron reemission was observed for all implantation energies (Fig. 2) indicating that even the most deeply implanted positrons drifted back to the exit surface. Thus they had a similar reemission probability as those implanted at lower energies. The experimental data are not consistent with a surface effect, which would have had a greater effect on the more shallowly implanted lower-energy positrons since more of these are able to diffuse back to the surface. A related field-induced phenomenon was observed by Gullikson and Henke[23] in x-ray-induced secondary electron emission, where enhancement was observed resulting from positive charging of solid xenon films. This was attributed to the electrons being drifted toward the surface by the electric field. A similar effect occurs here for the implanted positrons.

The effect of the electric field on the energy of the diffusion positrons (i.e., "heating" of the positron distribution) could be investigated using this model. This was observed as a reduction in the thermalized fraction and was significant at the field strengths used in this study. At high fields (>23 kV mm^{-1}) the fraction thermalizing was less than 1% at all implantation energies. Despite the heating of the positrons, which had the effect of increasing the probability of emission once at the surface, drift in the electric field was still responsible for the e^+ reaching the surface. A reversal of the applied electric field (i.e., $+8$ kV mm^{-1}) resulted in a reduction of the predicted reemission at 10 keV implantation to approxi-

mately 4%. This was in qualitative agreement with previous β^+ moderation efficiency measurements[10] where an almost total reduction in positron emission was observed when the field was reversed after positive charging of the surface by bombardment with positive ions.

The gradual reduction in reemission yield observed above a maximum of about 7 kV mm^{-1} and up to the cutoff (Fig. 3) was also modeled by the simulation and shown to result from heating of the positrons above the positronium formation threshold and into the so-called Ore gap.[24] This was reflected by an increase in the positronium formation fraction yielded by the computer simulation which at the highest electric fields became the dominant positron annihilation process for all implantation energies. Such heating effects have also been observed in gaseous argon at comparable density-normalized electric fields.[25,26] The observed maximum enhancement occurs at 7 kV mm^{-1} corresponding to a density-normalized electric field of $\sim 3.9\times 10^3$ V m^2 kg^{-1}, which is similar to those fields that cause the positronium fraction to rise in dense gases.[26] This behavior is also in good quantitative agreement with previous observations using surface-charged solid argon β^+ moderators.[11]

After this gradual decrease a very rapid decrease in the reemitted fraction was observed. For the sample thickness shown in Fig. 3 this occurred at a field strength of around 23 kV mm^{-1}. The results deviate at this point from the computer simulation, which could not account for this decrease. This effect did not occur at a particular value of electric field strength or surface charge density, both of which should vary approximately inversely with film thickness. The fact that it did not occur at a particular surface charge density suggested that it was not entirely due to trapping at anion sites on the surface of the film. The exact nature of this process is not clear at this stage.

By modification of the computer simulation it was possible to model the process of β^+ moderation in a (transmission mode) solid Ar film. It could then be used to fit the data obtained in a previous experimental study of Ar moderation under the influence of various surface potentials (applied electric fields).[10] In this case the implantation profile becomes exponential in form thus replacing assumption (1) given earlier with

$$P(z)=\alpha \exp(-\alpha z)\ ,$$

where $\alpha=29\rho$ cm^2 g^{-1} and ρ is the density of the target, in this case argon.[27] All other parameters in the simulation remain the same, although of course the positrons are now emitted from the opposite side to that of entry. The number of β^+ entering the film was taken as half of those created in the source plus those backscattered from the source mount. In the experiment this source mount was copper, which has an approximate backscattering coefficient of 30%.[28] Positrons diffusing back to the source mount are assumed to annihilate. The depth of the film (d) was taken as the only fitting parameter in order to model the experimental data since this parameter was poorly determined in the earlier work.[10] All other parameters were the ones used or determined in the previous simulation. The experimental and predicted moderation efficiency is shown in Fig. 4 as a function of surface potential (and thus electric field) for a film depth $d=1.3$ μm. As can be seen the zero field and high field values are in good agreement although those measured at low field fall below theory. The reason for this discrepancy was probably surface contamination. The field dependence was obtained experimentally by allowing the surface potential to decay and then measure the positron yield at various times after charging. The low-field measurements were therefore taken at relatively long times after deposition. The last data point, which deviates most from that predicted, was taken over 3 h after deposition. Taking the reported base pressure of 4×10^{-9} Torr and using the same depth dependence of pressure and time as that for the Ar film this would correspond to an $\sim$50-Å-thick overlayer of contaminants. This would undoubtedly have had a serious detrimental effect upon the positron emission efficiency.

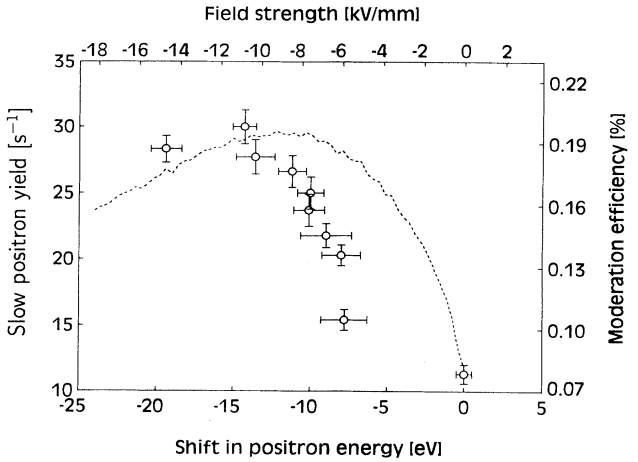

FIG. 4. Moderation efficiency for a solid argon β^+ moderator as a function of surface potential. The curve drawn is the result of the computer simulation mentioned in the text. For experimental details see Ref. 10.

VI. CONCLUSION

In summary, we have measured positron reemission properties of surface-charged solid argon for different thicknesses and different surface potentials and have obtained good agreement with a computer simulation from which a diffusion length of $1.7(+2.0,-0.4)$ μm and a mobility of $4.7(+2.9,-0.4)\times 10^{-3}$ m^2 V^{-1}s^{-1} have been derived. The value for the diffusion length is in disagreement with earlier studies, mainly due to a difference in the average energy loss per collision determined here to be $1.1(+0.4,-0.5)$ meV. The Monte Carlo simulation was also found to be in reasonable agreement with our previous studies of surface-charged Ar moderators. The results are consistent with a field-induced drift of the positrons in the bulk. A maximum enhancement in reemission was reached at an electric field strength of roughly 7 kV mm^{-1}. By increasing electric fields above 7 kV mm^{-1} a slow decrease in the

enhancement was observed; this was shown to result from heating of the positron energy distribution above the inelastic positronium threshold. Furthermore a rapid and almost total reduction of the reemitted fraction occurred for surface potentials above a thickness-dependent value in the range of -95 to -215 V. Further work is necessary in order to explain this latter effect.

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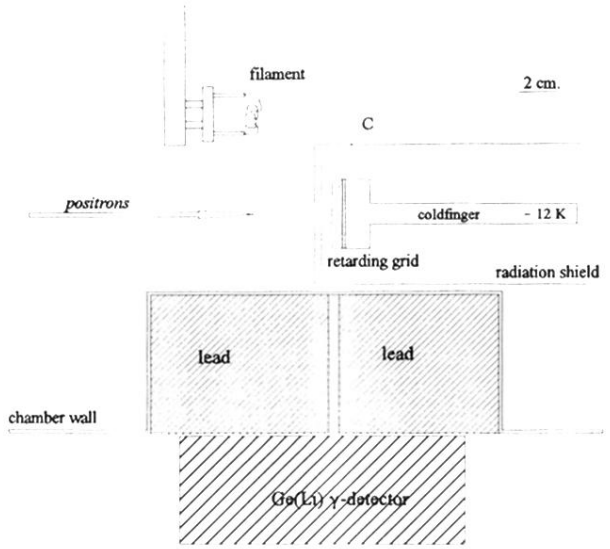

FIG. 1. The experimental setup near the sample. The sample copper backing *C* is electrically isolated from the cold finger.