Observation of Low-Energy μ^+ Emission from Solid Surfaces

D. R. Harshman[a] and J. B. Warren
Department of Physics, The University of British Columbia, Vancouver, British Columbia V6T 2A6, Canada

J. L. Beveridge, K. R. Kendall, R. F. Kiefl, and C. J. Oram
TRIUMF, Vancouver, British Columbia V6T 2A3, Canada

A. P. Mills, Jr., and W. S. Crane
AT&T Bell Laboratories, Murray Hill, New Jersey 07974

and

A. S. Rupaal and J. H. Turner
Department of Physics, Western Washington University, Bellingham, Washnigton 98225
(Received 31 January 1986)

Energetic positive muons were injected into the backs of thin moderators (lithium fluoride, quartz, and copper), and the transmitted beam energy analyzed. The μ^+ time-of-flight spectra for lithium fluoride and quartz were observed to have two components: a narrow distribution of slow (<10 eV) μ^+ [rate $\approx (1.6 \pm 0.35) \times 10^{-7}$ per incident μ^+ for lithium fluoride] and a broad fast component (0–2 keV), while the data for copper indicated only a fast contribution. Slow (<10 eV) negative muonium ions ($\mu^+ e^- e^-$) were also observed for the first time in vacuum.

PACS numbers: 71.60.+z

In this work we have investigated the energy distribution of low-energy muons (μ^+) transmitted through thin moderating solids. The approach taken draws guidance from analogous positron (e^+) experiments,[1] and may lead to a tunable beam of slow positive muons. Such a beam would be immensely useful from the standpoint of surface, atomic, and particle physics. For example, studies of muons and muonium atoms ($\mu^+ e^-$) on surfaces would provide information regarding surface magnetism and the hyperfine interactions of adsorbed muonium.[2] In addition, precision measurements of the optical excitation of muonium, as has been done for hydrogen[3] and positronium ($e^+ e^-$),[4] the search for muonium-antimuonium conversion,[5] and the formation of "muium" ($\mu^+ \mu^-$)[6] would be more feasible given a bright source of slow μ^+.

Slow positron beams are currently produced by utilization of negative-affinity metallic moderators. Such moderators are not expected to work for muons, however, because of the large μ^+-e^- correlation energy in a metal. Fortunately, ionic solids can also be used as slow-positron moderators[7] and a recent positron experiment[8] suggests that a large-band-gap solid may work as a slow μ^+ emitter as well. Of particular importance for surface emission processes are the atomic binding energy and the bulk diffusion rate (of the charged and neutral states) in the solid. Both the positron and the muon can capture an electron from the stopping medium to form the corresponding neutral species, positronium (Ps) and muonium (Mu), respectively. However, the binding energy of Ps is only $\frac{1}{2}$ that of Mu, making the energetics for μ^+ vs e^+ emission substantially different. As a result of the lighter mass of the positron ($m_\mu \approx 207 m_e$), the e^+ and Ps diffusion rates are typically much greater than those for μ^+ and Mu in the same material, whereas the μ^+ mean lifetime ($\tau_\mu \approx 2.197$ μs) is much greater than the lifetime of positrons in solids (typically <2 ns).

To study the energy spectrum of muons emerging from the surface of possible moderators, the apparatus shown in Fig. 1 was designed and constructed. It consists of a high-vacuum scattering chamber, an electrostatic extraction lens, and a magnetic spectrometer. Energetic (4.2 MeV, $\sim 2.5 \times 10^5$/s) muons, from the M20 secondary channel at TRIUMF, enter the scattering chamber through a water-cooled reentrant window assembly; the muons pass through an incident 0.25-mm scintillator, where they are detected, then through two 0.013-mm aluminum heat shields, and finally through a 0.025-mm stainless-steel vacuum window. The target is oriented with the surface normal at a 45° angle with respect to the incident muon beam. The electrostatic lens, immediately downstream from the target, collects and accelerates (to 10 keV) any low-energy charged particles that emerge from the target. The extracted particles are then injected into a dipole-quadrupole-quadrupole magnetic spectrometer which momentum selects and focuses the 10-keV charged particles onto a channelplate detector. A positron telescope array, subtending a total solid angle of about $8\pi/3$ sr, is situated around the channelplate detector to

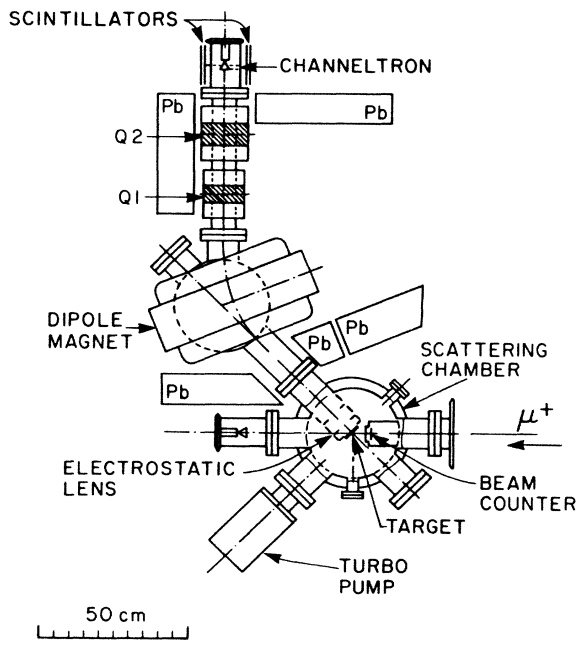

FIG. 1. Diagram of the apparatus.

detect the decay positrons. Assuming an efficiency of 50% for the channelplate detector for 10-keV muons, and requiring the observation of a decay positron, we estimate the experimental detection efficiency of the apparatus to be 10%, exclusive of the spectrometer acceptances.

Three targets were studied: air-cleaved lithium fluoride (LiF, 0.18 mm and 0.23 mm thick), z-cut quartz (SiO_2, 0.20 mm), and polycrystalline copper (Cu, 0.05 mm). The LiF and SiO_2 crystals were oriented with the $\langle 100 \rangle$ axis and c axis normal to the emitting surfaces, respectively. The incident beam momentum was adjusted to maximize the number density of muons thermalizing near the downstream surface of the targets. The experiments were performed in a vacuum of 5×10^{-8} Torr with the targets at an elevated temperature of about 150 °C. The purpose of the elevated temperature was threefold: to enhance diffusivity, to increase ionic conductivity (in the case of LiF), and to help maintain clean surfaces. Similarly treated samples of LiF have been shown to have less than a monolayer of surface contamination.[9]

Two time spectra were collected simultaneously: a time-of-flight (TOF) spectrum between single events in the incident μ^+ counter and the channelplate detector, and a μ-decay spectrum which was started on the channelplate-detector signal and stopped upon the detection of a muon-decay positron in the position telescope array. To reduce backgrounds, a triple coincidence was demanded for the TOF spectrum; any TOF events that were not associated with a subsequent μ-decay event were rejected. The correspondence of

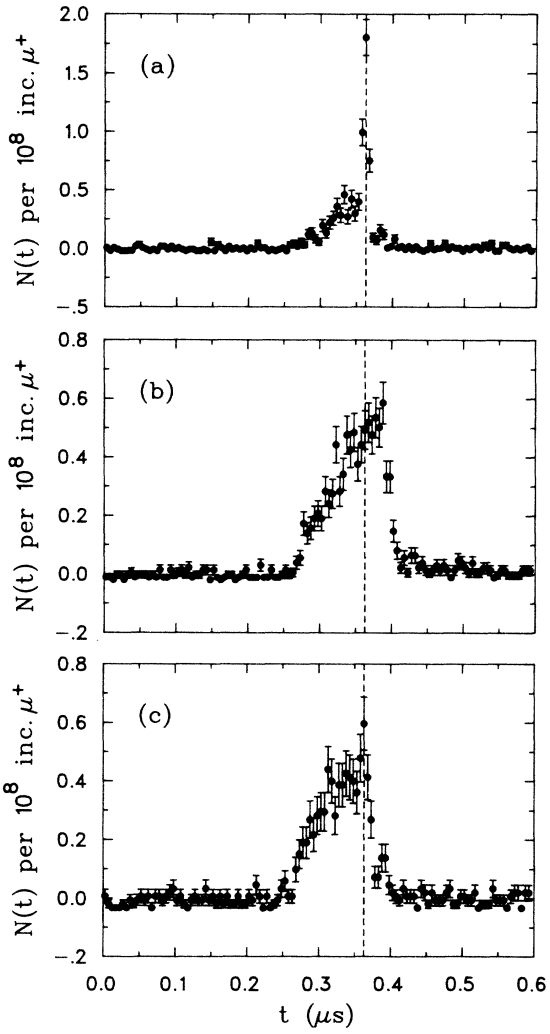

FIG. 2. Time-of-flight spectra for μ^+ from the surface of (a) LiF, (b) SiO_2, and (c) Cu. The data are normalized to 10^8 incident μ^+ and the errors shown are statistical.

the TOF through the spectrometer coupled with the associated muon-decay spectrum, provided a positive signature of the detected particles as muons. The experimental time resolution, determined by measurement of the TOF of beam positrons (30 MeV/c) between the incident muon counter and a channelplate detector mounted on the straight-through port of the scattering chamber, was estimated to be ~ 2 ns.

The TOF spectra taken with LiF, SiO_2, and Cu are shown in Fig. 2. A small flat background (typically <0.3 count/channel), determined from the integrated rate over a 100-ns interval between 75 and 175 ns, has been subtracted. The dashed line (t_0 at 363 ns) represents the "zero energy" position (i.e., the calculated average TOF for a prompt reemitted μ^+ having energy supplied solely by the extraction optics). Events at times $t > t_0$ are thus attributed to delayed

emission. The LiF data of Fig. 2(a) clearly show two components: a slow (<10 eV) μ^+ component and a broad higher-energy (or fast) background. The fast component was observed for all three targets and is attributed to beam muons degraded to 0–2 keV energies. The normalized integrated fast-μ^+ rates for LiF, SiO_2, and Cu, calculated over a 50-ns interval between 300 and 350 ns, are $(3.0\pm0.20)\times10^{-8}$, $(3.6\pm0.18)\times10^{-8}$, and $(3.9\pm0.29)\times10^{-8}$ per incident μ^+ respectively. The slow component, observed in LiF and (perhaps) SiO_2, is attributed to muons that emerge from the moderator surfaces with low kinetic energies (<10 eV, estimated from the width of the LiF peak). The LiF data exhibit a sharp cutoff of the slow events at late times ($t > t_0$), whereas the SiO_2 data of Fig. 2(b) show a much broader distribution, which appears also to be shifted by ~ 25 ns to later times, possibly a manifestation of charging. The normalized integrated slow-μ^+ rates, measured for LiF, SiO_2, and Cu, are $(4.5\pm0.25)\times10^{-8}$, $(4.6\pm0.20)\times10^{-8}$, and $(2.9\pm0.25)\times10^{-8}$, respectively. The integration was performed over a 50-ns time interval between 350 and 400 ns. By subtracting the Cu data from the LiF data and taking the aforementioned experimental efficiency into account, one obtains a slow-μ^+ flux of $(1.6\pm0.35)\times10^{-7}$ for LiF. Although not clearly evident from Fig. 2(b), the increased slow-μ^+ integrated rate from SiO_2 over Cu would suggest an enhanced slow-μ^+ emission similar to that observed for LiF.

Similar emission behavior has been reported[8] for positrons in the same materials; positrons of kiloelectronvolt energies, implanted into ionic single crystals, are observed to be subsequently reemitted with a continuum of energies having a maximum approximately equal to the band gap. Expanding upon an earlier suggestion,[10] an emission model was proposed[8] in which the e^+ is Auger emitted when the Ps electron falls into an acceptor level at the moderator surface. Assuming that the same mechanism(s) might also be involved in μ^+ emission, the maximum available energy would correspond to the Mu electron recombining with a hole at the top of the valence band. The actual energy available may be somewhat less than this, particularly in the case of salt-structure alkali halides which show evidence for a self-trapped hole (V_k center); for LiF, the energy required to form a hole center is 6.866 eV.[11] For Mu in LiF (band gap $= E_g = 13.7$ eV),[12] SiO_2 ($E_g \approx 9$ eV),[8] or Cu, this simple emission process is thus not energetically possible, unless it is coupled with a "recombination-assisted diffusion" model.[13] In this case, a Mu atom may act to catalyze electron-hole recombination, with the excess energy being converted to kinetic energy of the Mu atom. Later, the electron of the now "fast" Mu atom may combine with another hole which may, depending upon the available energy (kinetic+hole), cause the Mu atom to break up. If this process occurs near a surface, the muon could be expelled from the solid in a manner consistent with diffusion-controlled emission. An alternative suggestion is that the slow-μ^+ component is due to epithermal muons which fall below the band-gap energy; above E_g, energy loss occurs primarily through electron-hole pair production, whereas below E_g, energy is lost at a comparatively lower rate through phonon excitation. This process would also exhibit a strong correlation with the band-gap energy, but would not be strongly diffusion dependent.

Within the context of a diffusion-controlled model (assuming a random walk process), the slow-μ^+ TOF spectrum would exhibit a shape defined by

$$N(t)=\frac{1}{2}\left[F(T)y_0\frac{1}{r}\right]\left[\left(\frac{D}{\pi}\right)^{1/2}\frac{1}{\sqrt{t}}\exp\left[-\left(\lambda+\frac{1}{\tau_\mu}\right)t\right]\right]. \quad (1)$$

Here $F(T)$ is the bulk-formed Mu fraction at temperature T, λ is the rate of loss of bulk Mu, y_0 is the probability for μ^+ emission, $1/r$ represents the normalized stopping distribution of the incident μ^+ (where r is the range width in the material), and D is the "average" diffusion constant of the μ^+-Mu ensemble. The λ parameter is included to allow for the possibility that Mu undergoes a thermal transition to a diamagnetic state (μ^+ or Mu^- ion), as has been observed in some alkali halides.[14]

To look for evidence of diffusion-delayed emission the TOF data have been fitted in the range $t > t_0$ by use of Eq. (1). Because the time-resolution distribution exhibited no long tails, the effect of the resolution function was not considered important in the analysis. For LiF and Cu, the fits gave $\lambda = 63^{+41.5}_{-27.0}$ μs^{-1} and $\lambda = 73^{+53.4}_{-31.6}$ μs^{-1}, respectively. In the case of Cu, no such emission is expected, and the rather large λ value is consistent with this. On the other hand, the relatively large λ obtained for LiF implies either that the muons in the diffusing ensemble are indeed lost at a high rate, or that the emission is not diffusion controlled. Unfortunately, the possibility of distortions due to charging for SiO_2 makes the time dependence in this case somewhat suspect. Thus, it is at present not possible to draw any conclusions regarding diffusion-delayed processes.

Measurements were made also with the polarities of the electrostatic lens and dipole-quadrupole-quadrupole spectrometer reversed. The resulting TOF spectra, with the backgrounds (typically <0.05 count/channel) removed, are shown in Fig. 3. The data for both LiF and Cu clearly indicate a peak at t_0 (dashed line), whereas very few events were recorded in the case of SiO_2. This peak is comparable in width to the slow-μ^+ TOF peak of Fig. 2(a) and is attributed to

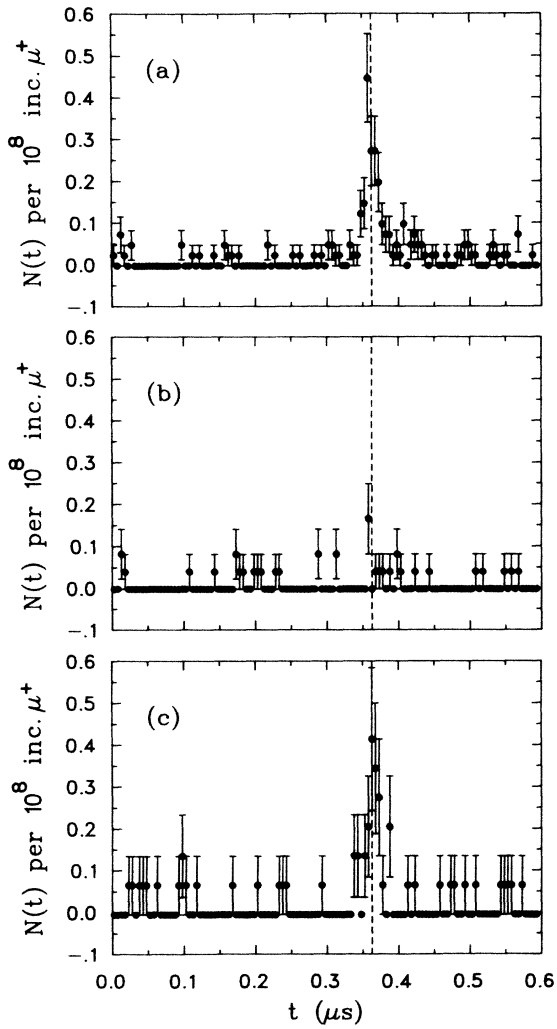

FIG. 3. Time-of-flight spectra for Mu^- ions from the surface of (a) LiF, (b) SiO_2, and (c) Cu. The data are normalized to 10^8 incident μ^+ and the errors shown are statistical.

Mu^- ($\mu^+e^-e^-$) ions emerging promptly from the target at low (<10 eV) kinetic energies. This charge state, which has heretofore not been observed in vacuum, is clearly shown here to be formed at very low energies as there is no evidence of a fast-μ^+ contribution. Accounting for the experimental efficiencies, the integrated slow-Mu^- rates for LiF and Cu are $(1.6 \pm 0.21) \times 10^{-7}$ and $(1.3 \pm 0.35) \times 10^{-7}$ per incident μ^+, respectively, while the data for SiO_2 indicate a much lower rate of $(0.38 \pm 0.145) \times 10^{-7}$. The reduction in Mu^- events observed for SiO_2 may be significant; however, given the possibility of charging, this will require further investigation.

In summary, low-energy μ^+ emission has been observed from LiF and (possibly) SiO_2, while no such emission was observed for Cu. In addition, Mu^- ions have been observed for the first time in vacuum. The observed emission for both charge states has been shown further to be a strong function of the target medium. Clearly, much work remains to be done before a usable beam of slow μ^+ (or Mu^-) can be realized. Of interest to future studies is the detailed measurement of the energy spectra for both charge states and a quantitative comparison of these results to the corresponding positron data. The effects of surface contaminants and structural damage on the energetics of emission would also be of interest. Additional information on the emission mechanism(s) involved could also be obtained by extending the measurements to rare-gas solids, such as neon ($E_g = 22$ eV),[12] as well as studying the temperature dependence of the flux ratio of the two charge states for various materials, adsorbates, and crystal orientations.

The authors would like to thank F. C. Hallberg of the U.S. National Aeronautics and Space Administration for providing the LiF crystals, J. Doornbos and G. S. Clark for their assistance with some of the apparatus design, and the TRIUMF staff for support.

[a]Present address: AT&T Bell Laboratories, Murray Hill, N.J. 07974.

[1]S. Berko and H. N. Pendelton, Annu. Rev. Nucl. Part. Sci. **30**, 543 (1980).

[2]D. R. Harshman, Ph.D. thesis, University of British Columbia, 1986 (unpublished).

[3]T. W. Hänsch, S. A. Lee, R. Wallenstein, and C. Wieman, Phys. Rev. Lett. **34**, 307 (1975).

[4]S. Chu and A. P. Mills, Jr., Phys. Rev. Lett. **48**, 1333 (1982).

[5]J. J. Amato, P. Crane, V. W. Hughes, J. E. Rothberg, and P. A. Thompson, Phys. Rev. Lett. **21**, 1709 (1968); K. R. Kendall, Ph.D. thesis, University of Arizona, 1972 (unpublished); G. M. Marshall, Ph.D. thesis, University of British Columbia, 1981 (unpublished).

[6]K. Nagamine, in *Proceedings of the Third LAMPF II Workshop, Los Alamos, New Mexico, July 1983,* edited by J. C. Allred, T. S. Bhatia, K. Ruminer, and B. Talley (Los Alamos National Laboratory, Los Alamos, N. Mex., 1983), p. 258.

[7]P. G. Coleman and J. T. Hutton, Phys. Lett. **85A**, 459 (1981).

[8]A. P. Mills, Jr., and W. S. Crane, Phys. Rev. Lett. **53**, 2165 (1984).

[9]T. E. Gallon, I. G. Higginbotham, M. Prutton, and H. Tokutaka, Surf. Sci. **21**, 224 (1970).

[10]K. F. Canter, P. G. Coleman, T. C. Griffith, and G. R. Heyland, J. Phys. B **5**, L126 (1972).

[11]P. E. Cade, A. M. Stoneham, and P. W. Tasker, Phys. Rev. B **30**, 4621 (1984).

[12]A. B. Kunz, in *Elementary Excitations in Solids, Molecules and Atoms,* edited by J. T. Devreese, A. B. Kunz, and T. C. Collins (Plenum, London, 1974), Pt. B, p. 343.

[13]A. M. Stoneham, in *Exotic Atoms,* edited by K. Crowe, J. Duclos, G. Fiorentini, and G. Torelli (Plenum, New York, 1980), p. 209.

[14]K. Nishiyama, Y. Morozumi, T. Suzuki, and K. Nagamine, Phys. Lett. **111A**, 369 (1985).