

Surface Analysis and Atomic Physics with Slow Positron Beams

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Positrons were predicted by the Dirac (1) theory of the electron and were discovered by Anderson (2) 50 years ago. Being the antimatter equivalent of electrons, positrons have precisely the same mass and spin as electrons, but precisely the opposite charge and magnetic moment (3). Since a positron and an electron can annihilate into γ -ray photons, the otherwise stable positron decays in

Positron annihilation offers a unique probe for detecting vacancies in metals (4), for measuring the Fermi surfaces of disordered alloys (5), and for observing the quantum chemistry associated with a particle of very light mass (6). It must also be mentioned that a positron and an electron can bind together to form a light atom called positronium, first observed in 1951 by Deutsch (7). The measure-

Summary. Recent advances in slow positron beam techniques are making it possible to study the interactions of low-energy positrons with gas molecules and solid surfaces and to measure the properties of free positronium atoms. New surface related results include the observation of surfaces with negative positron affinity and the thermionic emission of slow positronium atoms, low-energy positron diffraction measurements, and the sensitive detection of near-surface crystalline imperfections. Two recent successful experiments in atomic physics are the formation of the positronium negative ion and the optical excitation of positronium for high precision spectroscopy. Prospects for a positron microscope and the study of exotic antimatter systems such as the two-component Fermi gas are based on the imminent possibility of enormous increases in the brightness and instantaneous intensity of positron beams.

the presence of ordinary matter. Because energy-loss cross sections are high, positrons usually slow down to nearly thermal energies before annihilating, even though they are formed with million-electron-volt energies in beta-decay or pair production. The positron lifetime (10^{-10} second) and the total momentum of the annihilation photons give useful information about the environment in which the positrons have thermalized. It is therefore not surprising that the positron technique has been responsible for a number of interesting results in the study of condensed matter.

ment of the properties of positronium yields sensitive tests of the theory of electrons, positrons, and photons called quantum electrodynamics (8).

The study of the interactions of positrons with matter has received a big boost from a new technique based on an insight by Madansky and Rasetti in 1950 (9): positrons do not necessarily have to annihilate in the same material in which they are slowed to thermal energies. Given a source of energetic positrons, we should be able to extract beams of positrons from a nearby suitably chosen material surface with properties analo-

gous to those of the cathodes from which we obtain electron beams. An advantage of a beam of positrons is that one can investigate samples that are too thin to thermalize a significant fraction of the high-energy beta-decay positrons of the usual experiment. It is thus possible to study the unique interactions of positrons with surfaces under controlled conditions. A second advantage is that the unwanted radiations and chemical properties of the radioactive positron source are completely separated from the sample being studied. This permits us to investigate rare processes which would otherwise be masked by background events.

The discovery of slow positron emission from a solid under β^+ bombardment was made by Cherry (10) in 1958 and confirmed 10 years later by Costello *et al.* (11). Following these early experiments came a period during which slow positrons were used successfully in many experiments while the art of slow positron production steadily improved. A particularly important discovery (12) was that a surface coated with magnesium oxide converted 480-kilo-electron-volt end point cobalt-58 positrons to 2-eV positrons with an efficiency $\epsilon = 3 \times 10^{-5}$. This moderator provided the basis for the observation of the surface formation of positronium (13) and its excited state (14, 15) in 1975, experiments which represent the start of the present era of slow positron research.

While this early development took place without the benefit of any definite knowledge of the physical processes involved, the positron interaction with the surface was recognized as an important factor in the workings of a successful moderator (16). Finally, in 1978 experiments were performed in which monoenergetic positrons were used to bombard atomically clean surfaces in ultrahigh vacuum. These experiments demonstrated that positrons implanted in a clean single-crystal metal target can diffuse back to the surface and be emitted as positronium (17) or as free low-energy

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positrons (18). The positrons are emitted predominantly in a forward lobe (19) with a maximum energy whose value is interpreted as the positron negative work function of the surface (20). This new understanding has led to moderators with conversion efficiencies better than $\epsilon = 10^{-3}$. The resulting beam strengths of more than 10^6 positrons per second with nearly thermal energies are creating new possibilities for experimentation on surfaces and solids and for studying the atomic physics of positronium and positron-molecule scattering at low energies.

Slow Positron Moderators

Before discussing some of these new experiments, it is appropriate to describe the "anticathode" by means of which we are able to extract a beam of thermal positrons from a radioactive β^+ source. The requirements for such a fast-positron-to-slow-positron convertor are that it be made of a high-density material with a long diffusion length λ and a negative affinity ϕ_+ for positrons. Apart from geometric factors, the fast-to-slow positron conversion efficiency would be

$$\epsilon \approx y_0(\lambda/\Lambda)$$

where Λ is the mean penetration depth of the β^+ particles (21) and y_0 is the fraction of positrons that are ejected once they reach the surface. The positron diffusion length is typically about 10^{-5} centimeter and the penetration depth is about 3×10^{-2} gram per square centimeter for a 500-keV end point β^+ spectrum. Thus one would expect $\epsilon \approx 10^{-3}$, provided $y_0 \approx 1$.

Figure 1 shows the configuration of a slow positron moderator and radioactive source (22, 23). The moderator is a single crystal of copper that has been cleaned and coated with a submonolayer of sulfur by heating to about 800°C. It is necessary to use moderator materials with low defect concentrations because various defects such as vacancies (4), voids (24), and dislocations (25) are very effective positron traps and would reduce the positron diffusion length. The surface must be free of contaminants such as layers of carbon and oxides because these will also trap the positrons if they are amorphous (26). A submonolayer of sulfur forms naturally on the surface because of diffusion of bulk impurities, but this layer actually helps the positrons to leave the solid by making the positron affinity for the solid more negative. The radioactive source irradiates the copper surface

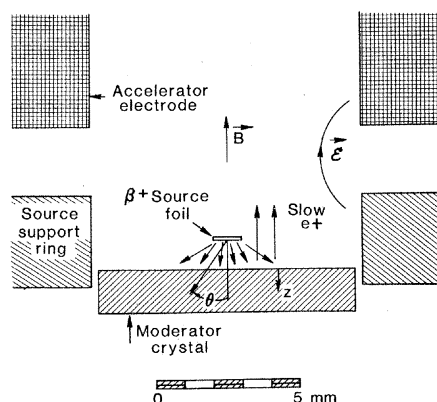


Fig. 1. Slow positron source consisting of a radioactive source foil and a clean single-crystal moderator (22). A portion of the energetic positrons ($\sim 10^5$ eV) from the β^+ emitter stop in the crystal. About 1 in 1000 of these diffuse back to the surface and are ejected from it with a kinetic energy (~ 1 eV) equal to the positron's negative affinity for the surface. The positrons form a beam that is transported to a target by a magnetic field. The beam kinetic energy may be varied from 1 eV to many kiloelectron volts by changing the potential on the moderator.

with energetic positrons, some of which diffuse to the surface after stopping in the metal. About half of the positrons that have reached the surface are emitted with energies characteristic of the -0.7 -eV positron affinity, and these are collected to form a slow positron beam. The efficiency $\epsilon = 10^{-3}$ can be improved somewhat by lowering the temperature of the copper crystal to increase the positron diffusion length (27). Using a ^{58}Co source with a strength of 0.5 curie (1.85×10^{10} disintegrations per second; β^+ emission probability, 14 percent; maximum β^+ energy, 475 keV), we obtain a beam of 2.5×10^6 slow positrons per second. Although this positron current of 0.4×10^{-12} ampere is small compared to typical electron beam currents, the positrons are easily detected because of their eventual annihilation and a number of interesting experiments are possible.

Positron-Surface Interactions

Some effort has been made to understand the slow positron emission process in more detail. For example, the angular distribution of positrons (19) has a narrow peak in the direction normal to a Cu(111) surface (see Fig. 2). This is what one would expect in a model where the positron is emitted by sliding down a potential hill whose height is $-\phi_+$, the positron's negative affinity for the copper. It is also found (28–30) that the

fraction y_0 of positrons that leave the solid increases with ϕ_+ as $y_0 = \exp(-\sqrt{\phi_0/\phi_+})$. For surfaces of copper and aluminum the constant has the value $\phi_0 = -0.27$ eV. The interpretation that the maximum energy of emission (0.8 eV in Fig. 2) is the positron's negative affinity or work function is strengthened by experiments (20) which show that the electron and positron work functions change by equal and opposite amounts when the surface dipole layer is changed by the addition of contaminants.

It was mentioned above that positrons are very effectively trapped by voids in metals (24). Thus from the viewpoint of Hodges and Stott (31), that a metal surface is a giant void, it is quite surprising that we actually do observe slow positron emission from metal surfaces. In fact, we now know that a positron surface state (32) does exist on many clean metal surfaces (33–35). Furthermore, it is also energetically favorable for a positron to leave the surface, taking with it an electron to form a free atom of positronium. In a typical case these three channels—slow positron emission, free positronium emission, and surface state trapping—represent approximately equally likely ways for a positron near a surface to be removed from the bulk crystal. A satisfactory microscopic explanation for the equality of the three transition rates is still lacking (30, 36).

Thermal Energy Positronium

It is fortunate that one of these three processes does not completely dominate the others because we are thus able to learn more about how a positron interacts with a surface. It is also fortunate that the binding energy of a positron surface state is such that we can thermally desorb the positron as a positronium atom without vaporizing the sample itself. In this process, which is analogous to the thermal desorption of hydrogen from a tungsten surface (37), the positron is initially in the lowest energy state $-E_b$ of its image potential well just outside the metal surface. Because of thermal agitation the positron can occasionally gain an energy of several times kT in motion parallel to the surface, where k is Boltzmann's constant and T is the sample temperature. The electrons of the metal are bound to it by at least an energy equal to the electron work function ϕ_- at $T = 0$. However, at finite temperatures the electrons can also have more energy because of thermal smearing of the Fermi distribution. Since the

energy to remove an electron is typically less than the binding energy of positronium, $\phi_- < \frac{1}{2}R_\infty = 6.803$ eV, the surface positron is likely to be ejected from the surface as a free positronium atom provided the energy deficit

$$E_a = E_b + \phi_- - \frac{1}{2}R_\infty$$

is made up by thermal fluctuations. At low temperatures the ultimate fate of a surface positron is to annihilate with an electron from the metal. Thermal desorption as free positronium will be significant only if the desorption rate $\Gamma = ae^{-E_a/kT}$ is comparable to the surface annihilation rate $\gamma \approx 2 \times 10^9 \text{ sec}^{-1}$. The preexponential factor a , intuitively the surface positron's normal velocity component divided by the dimensions of the surface well, can be shown (36, 38) to be $4kT/h$ in a model analogous to the Richardson-Dushman theory of electron thermionic emission. In the same model, the probability of positronium desorption has an Arrhenius temperature dependence (33–35) (see Fig. 3) from which we can deduce the activation energy E_a . From the known electron work function ϕ_- , surface state binding energies of about 3.0 eV are deduced for the surfaces of copper and aluminum (34). The E_b 's are much less dependent on surface orientation than are ϕ_- and E_a , which vary by as much as 0.5 eV. The calculation of the binding energy represents a considerable challenge (31, 39–43) because the light mass of the positron (44) invalidates the adiabatic approximation that is used to find the image potential of the much more massive proton (45).

Atomic Physics of Positronium

An exciting consequence of the discovery of thermionic positronium emission is that we now have a source of thermal positronium in vacuum for precision atomic physics experiments. The desorbed positronium velocities were shown to be thermal by a direct time-of-flight measurement (46) of the normal velocity component (see Fig. 4). The mean velocity is such that the positronium travels only about 2 cm during one mean lifetime of the triplet state, 1.42×10^{-7} second. It would therefore be possible to measure this important lifetime (47) in the absence of the buffer gas or confining walls that have been necessary until now.

The low velocities are also making it possible to induce optical transitions in positronium and to make precise measurements of the energy levels of this

unique atom. Even at thermal energies, however, the line broadening due to first-order Doppler shifts is excessive because of the small positronium mass. On the other hand, recent experiments to measure the $1S$ - $2S$ energy splitting in hydrogen (48) have exploited the first-order Doppler-free two-photon excitation technique (49) to achieve a very narrow line width. When the atom absorbs a photon from each of two counter-

propagating light beams, the first-order Doppler shifts are equal and opposite and hence cancel. There remains a second-order Doppler shift that for thermal positronium is only about one part in 10 million.

For a number of years it has been recognized that optical excitation of positronium would be desirable as the basis for a number of precision spectroscopic measurements of the positronium energy

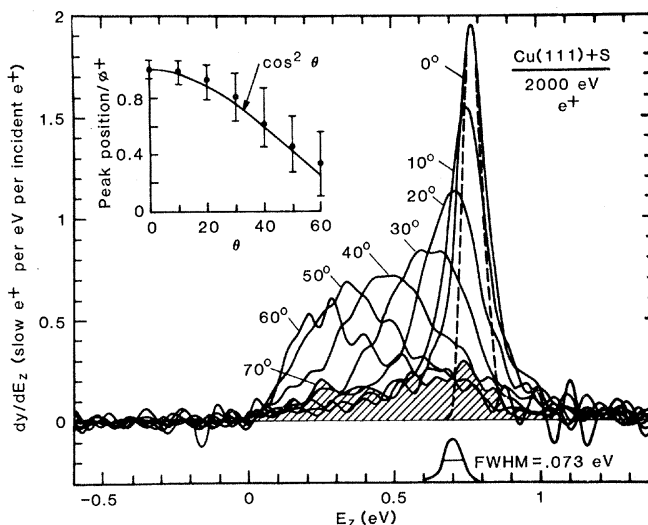


Fig. 2. Distribution of kinetic energies of positrons emitted by a copper surface bombarded with 1-keV positrons (19). The measurement yields the energy due to a single component of the positron momentum. As the angle θ of this component relative to the surface normal is changed, the peak of the distribution varies as $\cos^2\theta$ (inset). The interpretation is that the positrons are emitted moving perpendicular to the surface with an energy of 0.78 eV, equal to the negative of the positron work function.

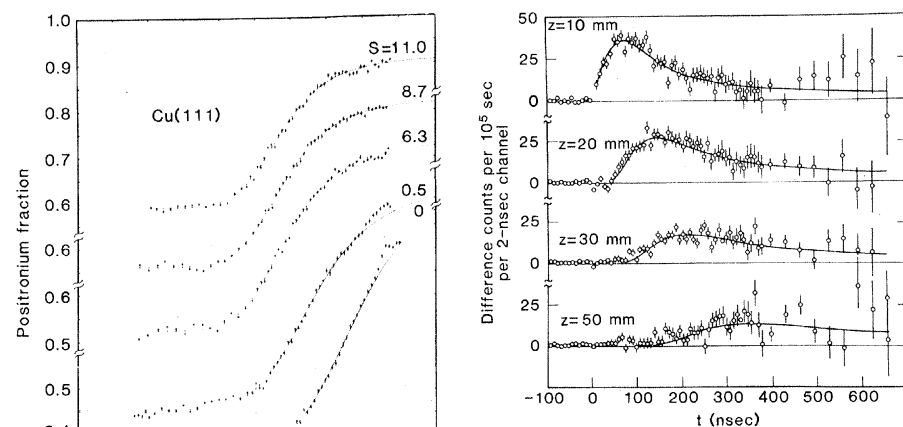


Fig. 3 (left). Positronium yield f of a copper single-crystal surface versus temperature T . The sample is bombarded with 30-eV positrons. The curves correspond to different

amounts of sulfur on the surface, ranging from an initially clean surface ($S = 0$) to a saturated coating of about one-third of a monolayer ($S = 11.0$). As the temperature increases the amount of positronium increases because of the thermal desorption of surface-bound positrons. Solid lines are curves with an Arrhenius temperature dependence $f = (f_0 + f_\infty a\gamma^{-1} \exp\{-E_a/kT\}) / (1 + a\gamma^{-1} \exp\{-E_a/kT\})$, that have been fitted to the data. The parameters f_0 and f_∞ are the positronium fractions at low and high T , respectively, and $a\gamma^{-1}$ is a rate constant. From the fitted activation energy, $E_a \approx 0.5$ eV, the binding energy of a positron in its image potential well at the surface is about 3 eV. The departure from the theoretical curves at $S = 0$ and $S = 0.5$ was presumably caused by diffusion of sulfur impurities to the surface as the sample temperature was increased. Fig. 4 (right). Time-of-flight measurements of the velocity of positronium thermally desorbed from a $\text{Cu(111)} + \text{S}$ surface at 1063 K (46). The positronium is detected in a region 10 mm wide centered a distance z above the copper surface. As z increases the time required for the positronium to reach this position increases, as expected. The positronium velocity is inferred to be about 10^7 cm sec^{-1} .

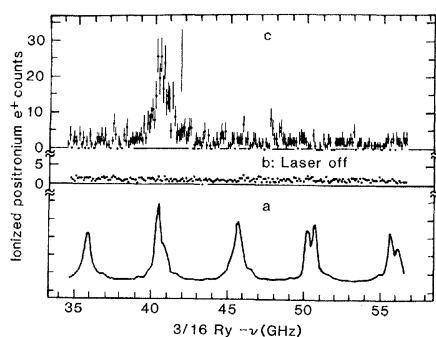


Fig. 5. Signal showing excitation of positronium from its ground state to its first excited state (51). The positronium was formed at a single-crystal copper surface (see Fig. 4). The large first-order Doppler shift in the resonance position is eliminated by driving the triplet $1S \rightarrow 2S$ two-photon transition with two counterpropagating laser beams at half the transition frequency of about $3/8 R_\infty$. As the laser frequency [measured by the Fabry-Perot fringes in (a)] sweeps through the resonance, an absorption signal (c) is seen which is not present when the laser is off (b). The resonance peak is shifted slightly from the predicted value (vertical line, $3/16 R_\infty$ plus quantum electrodynamic corrections) because of a Stark shift and uncertainty in the frequency scale (± 0.5 GHz).

level structure (50). This year, we have succeeded in producing enough positronium and laser light simultaneously to observe $1S$ - $2S$ transitions (51) analogous to the earlier hydrogen work of Hänsch *et al.* (52). The resonance line width in the preliminary experimental result shown in Fig. 5 is broadened by residual first-order Doppler shifts due to laser beam misalignment. Nevertheless, the measurement shows that the energy difference between the $n = 1$ and $n = 2$ triplet states of positronium is 1233608 ± 1 GHz. This value is 82 ± 1 GHz below what would be expected on the basis of the nonrelativistic Schrödinger equation, in agreement with the calculations of Ferrell (53) and Fulton and Martin (54). Figure 5 shows that there is sufficient signal to eventually measure the energy interval between $n = 1$ and $n = 2$ with a precision limited primarily by our ability to measure the laser frequency. It is especially interesting to know the positronium levels accurately because this atom is made of constituents that are believed to be described very well by the theory of quantum electrodynamics (8). Most other atoms have a less well understood nucleus and hence there are small but significant uncertainties in the theoretical predictions.

Other atomic physics experiments with slow positron beam techniques are the study of positron scattering by mole-

cules (55), measurement of the positronium lifetime in vacuum (47), and investigation of the excited states of positronium (14, 15, 50). In a recent experiment (56) it was also possible to produce the positronium negative ion ($e^+e^-e^-$) by a beam-foil technique similar to that used in making H^- ions from a proton beam (57). A 50-angstrom-thick grid-supported carbon film was bombarded with a beam of 500-eV positrons in vacuum. Some of the positrons passed through the film, and by analogy with the proton experiments one would expect some of these positrons to emerge bound to one or two electrons. Indeed, negative ions were detected by accelerating them to several thousand electron volts and observing the Doppler-shifted γ -rays (see Fig. 6) from the annihilation in flight of the fast ions. The existence of the positronium negative ion was first demonstrated theoretically by Wheeler (58), and recent calculations (59, 60) show that its binding energy relative to a positronium atom and a free electron should be 0.32667 eV. There are a number of interesting experiments yet to be done with the negative ion, including photoionization to produce fast positronium beams and exploiting the tunable γ -rays (Fig. 6) as a precision calibration linking γ -ray wavelengths (61) to the voltage standard.

Positrons for Surface Analysis

There is no doubt that studies of positron atomic physics and surface interactions are of fundamental interest. There are also practical aspects of positron beams that are beginning to be exploited for surface diagnostics. The well-established positron technique (4) for detecting vacancies and other crystalline imperfections in the bulk has recently been applied to the near-surface region by Lynn (26) and by Triftshäuser and Kögel (62). These investigators irradiated samples with monoenergetic positrons whose penetration depth into the solid can be varied from a few tens of angstroms to thousands of angstroms. If the positrons encounter crystalline defects they become trapped in the regions of lower ion core density. The result is a change in the positron diffusion constant and changes in the annihilation lifetime and γ -ray energy distribution that can easily be observed. Using this new technique, Lynn observed the annealing of an extremely thin (5 Å) oxide layer on an aluminum single-crystal surface (see Fig. 7). In a complementary development, Triftshäuser and Kögel demonstrated

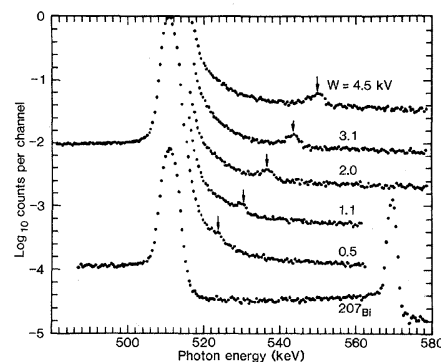


Fig. 6. Energy spectra of annihilation photons from 500-eV (56) positrons striking a 50-Å-thick carbon film. Behind the film is a grid at a potential $W = 0$ to 4.5 kV relative to the film. Positronium negative ions formed when some of the positrons pass through the film are accelerated by this potential and acquire a velocity in the direction of the Ge(Li) γ -ray detector. When the positron annihilates with one of the electrons in the ion, the result is an unmistakable blue-shifted annihilation photon. The position of these new γ -ray peaks relative to the large unshifted annihilation line at 511 keV moves to higher energy as W is increased. Arrows show the expected line centers. The ^{207}Bi line is shown for calibration.

that it is possible to use monoenergetic positrons to measure the depth profile of ion beam damage thousands of angstroms below a sample surface.

Since positrons have the same mass as electrons, they will exhibit similar diffraction effects when scattering from a single-crystal surface. Thus positron diffraction, recently demonstrated in the laboratory by Rosenberg *et al.* (63, 64) (see Fig. 8), may become a useful surface analysis technique, complementing the well-established methods of low-energy electron diffraction (LEED). The advantages of using positrons are that they interact with the solid in a simpler way because there are no exchange forces and that they are repelled from the ion cores, which attract and strongly influence low-energy electrons. Since the interpretation of LEED requires extensive computations, these simpler aspects of the positron may prove to be valuable (65-67).

Currently, however, positron diffraction cannot be called a practical technique because of the limited intensity of slow positron sources. The requirement for a LEED experiment is a particle beam with an energy E of 25 eV, a cross section d less than 1 millimeter, and an angular divergence θ less than 0.01 radian. The slow positron source described above (see Fig. 1) is about 10 mm across and has an intrinsic energy and angular divergence of about 0.25 eV and 1 radi-

an. By Liouville's theorem, the quantity $\theta d\sqrt{E}$ is conserved in the absence of dissipative forces. Thus we can form a 25-eV positron beam with a divergence of 0.01 radian, but the beam will be at least 100 mm across. This beam must be passed through a 1-mm aperture to achieve the desired characteristics, and there will be a 10,000-fold loss in intensity. Starting with about 10^6 slow positrons, we would thus have a beam strength of only 100 particles per second. Even though we can detect the diffracted positrons with about 50 percent efficiency with a microchannel electron multiplier array, the signal expected on the basis of Fig. 7 would be only one count every 10 seconds. (The data of this figure were obtained with a beam having much less restrictive θ and d .)

Intense Positron Beams

In order to improve the beam characteristics we must evidently not rely on conservative forces. Rather, the initial beam can be accelerated to, say, 10 keV and brought to focus in a 0.1-mm spot on a second slow positron moderator. Since the energy is small compared to beta-decay end point energies, the implantation depth of the positrons will be small and the secondary moderator efficiency will be very high, about 30 percent. These remoderated slow positrons can, in principle, be formed into the desired beam for diffraction studies with no further loss in intensity. Implementing this beam brightness enhancement technique (68) will require either fabricating transmission mode moderators from very thin clean single crystals (69) or using complicated electron optics to extract the secondary beam from the surface of a thick crystal.

It is of some importance to learn how to make the thin moderators because of the breadth of possible applications. For example, the brightness-enhanced positron diffraction beam described above could be reaccelerated to 10 keV and brought to a focus with a 10^{-4} cm diameter on a sample. One could scan the sample with this beam to obtain microscopic information about defect concentrations, grain boundaries, Fermi surface variations, and so on. There is also the possibility of using several stages of brightness enhancement in series to obtain a positron beam with a spatial resolution comparable to that of an electron microscope. In analogy to the methods established for scanning electron microscopy, the positron microscope signal

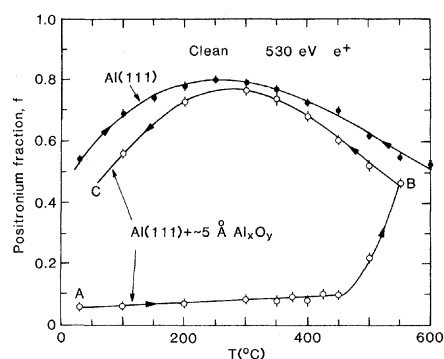


Fig. 7 (left). Positronium fraction f versus sample temperature T for clean and O_2 -exposed aluminum bombarded with 530-eV positrons. For the clean sample, as T is increased more positronium forms at first because of the thermal desorption of surface positrons. At higher temperatures f falls again because thermally generated vacancies in the aluminum start to trap the positrons, preventing them from reaching the surface where the positronium forms. The oxide-coated surface at first (point A) shows very little positronium formation because nearly all the positrons are trapped in the amorphous oxide layer. When the sample is heated to 550°C (point B) the oxide apparently crystallizes, permitting positrons to escape to the surface. On cooling ($B \rightarrow C$) the temperature behavior is similar to that of the clean surface. [From Lynn (26)]

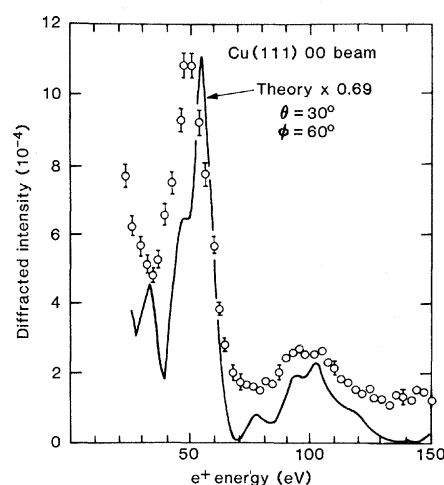


Fig. 8 (right). Intensity of the specular beam for positrons reflected from a copper sample (63). The theoretical prediction is from Read and Lowy (67).

could be obtained from a number of different effects in which the positron is sensitive to a specific surface or crystalline environment. In particular, the Doppler width of the annihilation γ 's, the positron and positronium reemission probabilities, the secondary electron current, and the desorbed ion current are all possible signals that would be sensitive to near-surface contaminants, defects, and composition.

In the near future, we can expect to see significant increases in the total intensity of positron beams as well as in their intrinsic brightness. First, we may obtain higher fluxes of slow positrons by using cascade shower positrons produced at the beam dump of a 10- to 100-MeV electron accelerator such as a microtron or linac (11, 70). The efficiency of conversion of fast electrons to slow positrons is easily estimated to be about 10^{-6} . Thus a 1-milliampere electron beam would give us 10^{-9} A of slow positrons, at least 1000 times more than we now have. Radioactive sources may also be made larger. The positron emitter ^{64}Cu is available with activities thousands of times higher than the ^{58}Co sources presently in use. Such higher fluxes will, for example, allow surface studies using the well-established positron techniques to measure electron momentum distributions in bulk crystals (71).

One effect that has not yet been mentioned here is the time-bunching of particle beams (72). We can trap and store in a magnetic bottle all the positrons that

have been emitted over a period of time about 10^{-4} second long. At some later time the positrons can be ejected from the bottle all at once and made to arrive within 10^{-8} second of each other at a target (73). Qualitatively new features of the positron interaction with surfaces arise when we think of combining this time-bunching with new large brightness-enhanced positron sources. Starting with, say, 10^{10} slow positrons per second and a better buncher, we can capture 10^{-3} -second portions of the beam to make 10^{-9} -second-long pulses containing 10^7 positrons each. Now we can imagine two stages of brightness enhancement after which we could have some 10^6 positrons arriving at a target at a single spot 10^{-5} cm in diameter. For the next 10^{-9} second before everything annihilates we could study a many-positron plus many-electron system. One might expect to see an e^+e^- plasma (74) and positronium molecules (75) or droplets (76). It might be possible to make the positron pulse coincide with an antiproton pulse and so make the first antiprotons. Letting our imagination run freely, we might even think about the annihilation γ -ray laser suggestion of Bertolotti and Sibilia (77).

Conclusion

From a modest beginning in 1958, slow positron beams have grown in steady strength by 10^5 and in instantaneous strength by about 1 billion. These beams

are being investigated as surface analysis probes and are making possible a variety of studies in atomic physics. In the near future we expect a further gain in steady intensity by as much as 10^4 and brightness enhancement of the flux per unit area by an additional factor of 1 billion. It seems likely that such enormous gains will be accompanied by an increasing array of applications and exciting new possibilities for fundamental experimentation.

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