LA-UR- 8 6 - 2 1 5 8 $CONF - 8\times y, 1384--3$ MASTER

TITLE: MUON CHANNELING IN Ge: EVIDENCE FOR PIONIUM FORMATION

RECEIVED BY   JUL 0 7 1986

AUTHOR(S): G. Flik, J. N. Bradbury, D. W. Cooke, R. H. Heffner,
M. Leon, M. A. Paciotti, M. E. Schillaci, K. Maier,
H. Rempp, C. Boekema, J. J. Reidy, H. Daniel.

DISCLAIMER

Los Alamos
Los Alamos National Laboratory
Los Alamos, New Mexico 87545

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

MUON CHANNELING IN Ge: EVIDENCE FOR PIONIUM FORMATION

G. Flik, J. N. Bradbury, D. W. Cooke, R. H. Heffner, M. Leon, M. A. Paciotti,
   and M. E. Schillaci, Los Alamos National Laboratory, Los Alamos,
   New Mexico 87545
K. Maier and H. Rempp, Max-Planck-Institut für Metallforschung and
   Universität Stuttgart, FRG
C. Boekema, San Jose State University, San Jose, California 95192-0106
J. J. Reidy, University of Mississippi, University, Mississippi 38677
H. Daniel, TU München, 8046 Garching, FRG

In a recent experiment observing muon channeling from the
decay of positive pions implanted in high-purity Ge single
crystals, the pion decay site is found to be sensitive to the
concentration of excess charge carriers produced by photon
absorption. Distinctly different channeling profiles are
observed in Ge at 100 K for illuminated samples (high carrier
concentration) as compared to unilluminated samples (low
carrier concentration). From these data we conclude that
different electronic states of the pion in a semiconductor
host lattice must account for the observed changes in the
pion location. Furthermore we suggest that the pion exists
in the solid both as the bare entity π^+ and as pionium
(π^+e^-), i.e., a hydrogen-like atom with a mesonic nucleus.

1. INTRODUCTION

The behavior of hydrogen, its heavier isotopes (d,t) and the lighter
hydrogen-like particles [positive muons (μ^+) and positive pions (π^+)] in
semiconductors is of considerable interest. A number of experimental techniques
(e.g., ion-beam channeling /1/, muon spin relaxation /2/, infrared and Raman
spectroscopy /3/) have been applied to this problem, but as yet no consistent
microscopic picture has emerged to describe the observed interaction of hydrogen
and its isotopes in semiconductors. Using the new technique of muon channeling
from implanted positive pions /4/, we have investigated the behavior of these
pions in single crystals of pure and doped Ge. These experiments were conducted
at the Biomedical Channel of the Clinton P. Anderson Meson Physics Facility
(LAMPF), and at the Swiss Institute for Nuclear Research (SIN).

2. EXPERIMENTAL

Positive pions, which have 1/7 the mass of protons and a 26 ns lifetime,
were implanted into single-crystalline Ge where they decayed according to $\pi^+ \rightarrow \mu^+$
$\rightarrow \nu_\mu$, yielding muons with 4.12 MeV energy. A small fraction ($\sim 10^{-5}$) of these
muons undergo channeling along the <110> crystallographic direction giving rise
to an angular distribution depending on the π^+ decay site. At a distance of 12.5
m from the sample the muon angular distribution was monitored by a two-
dimensional, position-sensitive scintillation detector with a spatial resolution
of 1.8 cm (yielding an angular resolution of 0.083°) and an energy resolution of
390 keV. This latter quantity is important for the following reason: with
increasing depth of pion implantation into the sample an increasing fraction of
initially channeled muons will be dechanneled /5/, i.e., they will lose the
information defining their point of origin as a result of multiple scattering
with electrons and host nuclei. These unwanted dechanneled muons were separated
from the channeled ones by energy analysis.

Ge single crystals were oriented such that their <110> axes pointed toward the center of the detector. In order to produce a large concentration of excess charge carriers without introducing impurities that might act as pion traps (as in a heavily doped sample), we directed the radiation from a high-intensity tungsten lamp onto the sample surface. Taking advantage of the time structure of the LAMPF pion beam (pulse repetition period 8.3 ms, pulse length < 750 μs), the light was chopped so that each light pulse overlapped alternate pion pulses striking the crystal. This technique resulted in a high carrier concentration when the light was on and, because of rapid surface and bulk recombination, an intrinsic concentration when the light was off. The procedure allowed us to 1) observe the effects of different carrier concentrations at the same sample temperature, 2) reduce the heat load on the sample, and 3) minimize the effects of any small shift in crystal orientation or drifts in electronics with time.

Most of the photons produced by the tungsten lamp (T ≈ 3400 K) and transmitted through the lucite light guide have energies exceeding the bandgap for Ge (E_g = 0.67 eV). Upon illumination, carriers are produced at the surface within a depth $\alpha^{-1} \approx 1$ μm, α being the absorption coefficient, whereupon they rapidly diffuse into the bulk. For typical bulk and surface recombination times /6/, this diffusion distance far exceeds the 50 μm depth from which the channeled muons emanate. We conclude that within this depth the carrier concentration resulting from illumination is constant, a typical value being $10^{15\pm1}$cm^{-3} for Ge.

3. RESULTS AND DISCUSSION

In Fig. 1 we show angular distributions of the relative muon channeling yields for Ge as a function of the angle ψ measured with respect to the <110> direction. Data are presented for light-on and light-off conditions at two different temperatures. Variation in the sensitivity of the detector over its surface was removed by dividing the channeling data at each point on the detector by "random" data where the channeling peaks are completely washed out by the scattering of a thin gold foil placed between the sample and the detector. The muon channeling distributions are expected to be symmetrical about the <110> direction, and the measured muon distributions are found to be symmetrical within errors. Hence an azimuthal average was made about the <110> direction, yielding the angular distributions in Fig. 1. The plots represent the ratio of channeled-muon intensity to nonchanneled intensity at each angle ψ within an energy interval at the maximum muon energy.

At 100 K there are striking differences between the muon angular distributions (channeling profiles) taken with light off and light on [compare Figs. 1(a) and 1(b)]. For example, with light off we see both a central peak (i.e., ψ = 0°) and an off-center one (ψ = 0.2°), whereas with light on we observe only an off-center peak (ψ = 0.1°). At 200 K [see Figs. 1(c) and 1(d)] we observe only slight differences between the light-on and light-off conditions, each profile exhibiting a central peak with hints of possible off-center peaks. We attribute the differences in the muon-channeling profiles observed at 100 K to the occupation of <u>different pion sites</u> resulting from an increased carrier concentration during the light-on condition. Furthermore we note that the pion site (or sites) at 200 K is different than at 100 K, and is essentially unaffected by light. At 200 K, thermal excitation increases the free carrier density by ~10^8 as compared to 100 K, thus making insignificant any increased carrier concentration due to light on.

For a qualitative interpretation of the data in terms of specific site occupancy we refer to Fig. 2, which represents the projection of a diamond lattice onto a (110) plane. The position of hexagonal (H) and tetrahedral (T) sites as well as antibonding (AB) sites are shown with respect to the <110> channel. Figure 1 illustrates muon-flux enhancement, i.e., channeling rather than blocking /7/, so one can immediately exclude substitutional sites as candidates for possible pion locations. Moreover, channeling theory /8/ predicts that pion sites located at or very near a channel center produce muon channeling peaks centered at ψ = 0, whereas off-center sites yield off-center (i.e., ψ > 0)
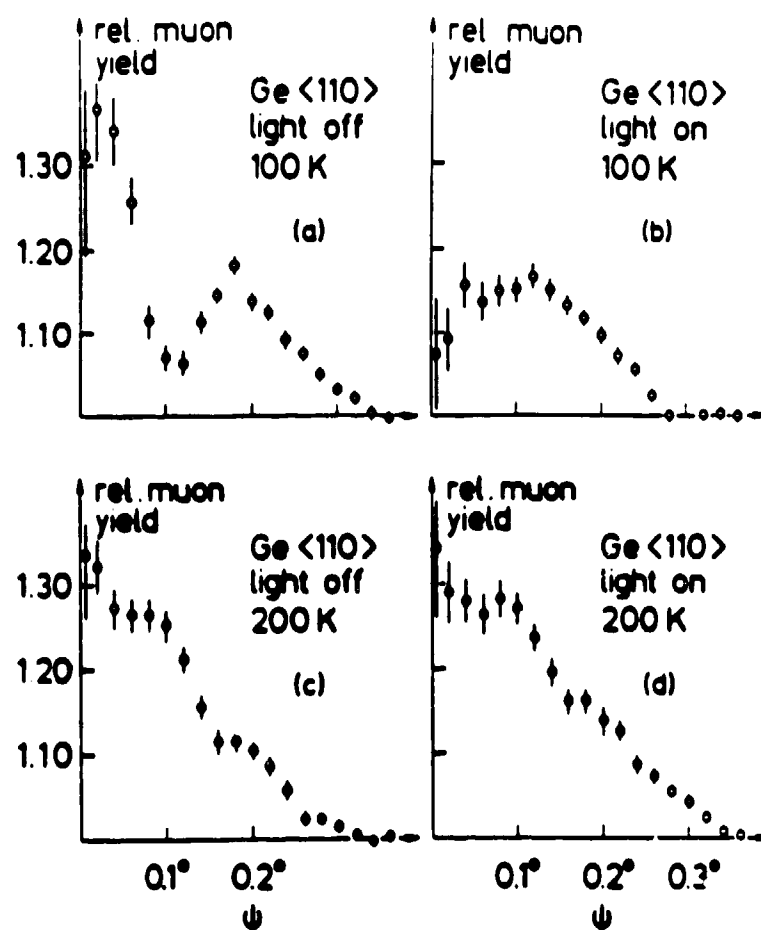
Fig. 1. Argular distributions of relative muon channeling yields for Ge as a function of the angle ψ measured with respect to the <110> direction. The data correspond to the following experimental conditions: (a) light off at 100 K, (b) light on at 100 K, (c) light off at 200 K, and (d) light on at 200 K.
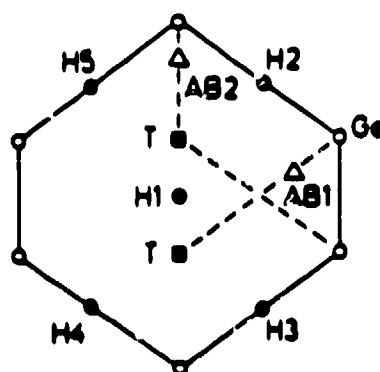

Fig. 2. Projection of a diamond lattice onto the (110) plane. Hexagonal (H1-H5), tetrahedral (T), and antibonding (AB) sites are illustrated.

peaks. Unfortunately, theoretical efforts have shown that one cannot resolve the T and H1 sites in a muon-channeling experiment. Thus we cannot presently make an unambiguous correlation of observed channeling peaks with specific pion sites. Future work will, however, include muon channeling profiles taken along <100> and

produces muon-flux enhancement for H sites but diminution (i.e., blocking) for T sites /5/, such data should be useful in differentiating between the two. Nevertheless we discuss the various possibilities for pion sites in Ge based upon the present data.

If H-site occupancy is preferred when the light is off then the central peak could be due to a pion residing at H1 whereas the off-center peak could be attributed to H2-H5 occupancy. For light on, the pion could preferentially occupy the T site. This interpretation seems plausible based upon the angular position of the off-center peak for Ge during light on. That is, we note that the off-center peak occurs at a smaller angle for light on than for light off. One must be somewhat careful with this conclusion, however, since it might be possible to generate the off-center channeling peak in Fig. 1(b) by appropriately weighting the central and off-center peak of Fig. 1(a). This possibility, coupled with our lack of knowledge regarding T and H site resolution, suggests a second, equally plausible, interpretation of the observed muon-channeling profiles. The central peak observed in Fig. 1(a) might be associated with pions occupying H1 and/or T sites, while the off-center peak represents AB site occupancy. With light on [Fig. 1(b)], a larger portion of the pions occupy AB sites than H1 and/or T sites thus yielding an off-center peak. At 200 K Ge exhibits a rather broad central peak for both light on and light off, possibly indicating multiple pion site occupancy.

Regardless of the uncertainty in specifying pion sites from the observed channeling profiles, it is clear that for certain temperature intervals a pion site change occurs when the carrier concentration is increased (light on). This effect can be understood qualitatively in terms of different electronic states of the pion. From muon spin relaxation (μSR) experiments, where positive muons (μ^+) are implanted into semiconductors /2,8/, it is known that muons form different electronic states, viz., diamagnetic (bare) μ^+, "normal" muonium (Mu = μ^+e^-) with a spherically symmetric electron distribution, and "anomalous" muonium (Mu*) with an anisotropic electron distribution, possibly resulting from bond formation. We suggest that normal pionium (Pi = π^+e^-) and anomalous pionium (Pi*), analogs of the muon states, are formed in Ge. We should expect bare pion (π^+) states as well; however, it seems highly unlikely that a bare pion would occupy different sites due to increased charge-carrier concentration.

Hartree-Fock and Hückel calculations /9,10/ demonstrate that the lattice potential, E_T^*, for 1s hydrogen isotopes at T sites is an absolute minimum. Thus, formation of Pi should occur at T sites, a location consistent with our data. Furthermore, it is known that the lattice potential, E_H^*, of 1s hydrogen isotopes occupying H sites represents an absolute maximum, with $E_H^*-E_T^*$ = 1.2 eV for atomic hydrogen in silicon /9/ and $E_H^*-E_T^*$ = 0.8 eV for Mu in diamond /10/. Therefore, low-symmetry H sites should be unstable for Pi. Thus, H-site occupancy is possible only if pionium forms bonds with each nearest-neighbor Ge atom (i.e., Pi*). Similarly, pionium at an AB site is expected to be chemically bonded with the nearest-neighbor Ge atom with which the AB site is associated. As H and/or AB sites are suggested by our data, we conclude that pionium states exist which cannot be solely explained by formation of Pi at T sites. We associate these states with anomalous pionium (Pi*).

In a related experiment recently conducted at SIN, we measured the relative muon yield as a function of temperature for both n-doped (Sb, 1.5×10^{15} cm^{-3}) and p-doped (Ga, 1.5×10^{15} cm^{-3}) Ge. The results are shown in Fig. 3, where the muon yield corresponds to the area under the channeling peak. For p-doped Ge the yield is nearly temperature independent for 25 < T < 200 K, whereas the n-doped sample exhibits significant temperature dependence. This means that the pion site is different for p- and n-doped Ge. The U-shaped trend of the muon yield data for n-doped Ge agrees with the light-on results for undoped Ge (LAMPF). This further supports our conclusion that excess electrons influence the pion site.

To summarize, the muon channeling results presented herein, specifically the observation of a pion site change, taken together with the theoretical results /9,10/, strongly indicate the existence of both normal pionium and anomalous
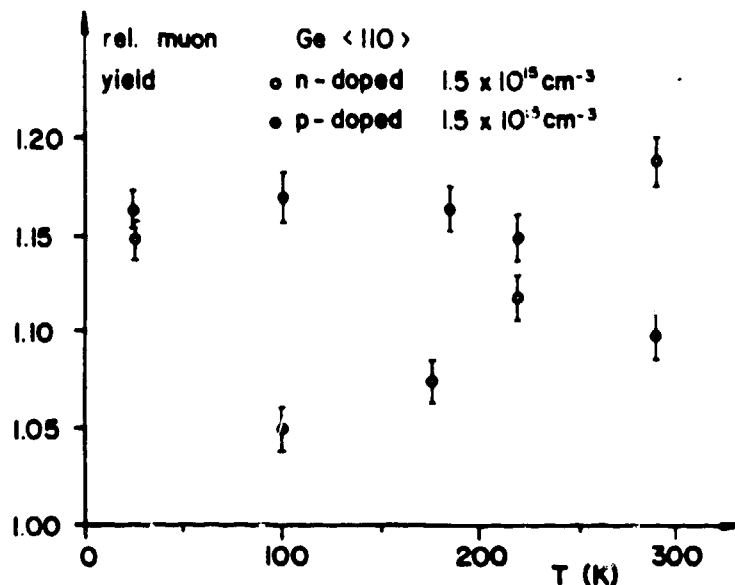
Fig. 3. Relative muon yield for n- and p-doped Ge.

pionium in Ge. To our knowledge, this is the first evidence suggesting the formation of pionium in a solid.

ACKNOWLEDGEMENTS

REFERENCES

/ 1/   S. T. Picraux and F. L. Vook, Phys. Rev. B18(1978)2066.

/ 2/   B. D. Patterson, in Muons and Pions in Materials Research, eds. J. Chappert and R. I. Grynszpan (Elsevier, New York, 1984) p. 161.

/ 3/   B. N. Mukashev, K. H. Nussupov, M. F. Tamendarov, and V. V. Frolov, Phys. Lett. 87A(1982)376.

/ 4/   K. Maier, G. Flik, D. Herlach, G. Jünemann, H. Rempp, A. Seeger, and H. D. Carstanjen, Phys. Lett. 83A(1981)341.

/ 5/   H. D. Carstanjen and A. Seeger, in Muons and Pions in Materials Research, eds. J. Chappert and R. I. Grynszpan (Elsevier, New York, 1984) p. 293.

/ 6/   L. P. Hunter, in Handbook of Semiconductor Electronics (McGraw-Hill, New York, 1962).

/ 7/   G. Gemmell, Rev. Mod. Phys. 46(1974)1.

/ 8/   A. Weidinger, G. Balzer, H. Graf, E. Recknagel, and Th. Wichert, Phys. Rev. B24(1981)6185.

/ 9/   V. A. Singh, C. Weigel, J. W. Corbett, and L. M. Roth, Phys. Stat. Sol. (b)81(1977)637.

/10/   N. Sahoo, S. K. Mishra, K. C. Mishra, A. Coker, T. P. Das, C. K. Mitra, L. C. Snyder, and A. Glodeanu, Phys. Rev. Lett. 50(1983)913.