

the present work.

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of  $n_{\text{eff}}$  corresponding to  $0.5 < \hbar\omega < 3.5$  eV. Uncertainties of absolute values of  $\sigma(\omega)$  are estimated to be  $\pm 10\%$ . Relations between optical terms used throughout the text are

$$N^{-1} \int_0^\infty \sigma_{\text{ib}}(\omega) d\omega \propto (m^* - m_0)/m^* = n_{\text{eff}} \\ \propto \int_0^\infty [M(\omega) \cdot J(\omega)/\omega] d\omega$$

[ $N$  = atom and conduction-electron density,  $M(\omega)$  = average value of squared transition matrix elements].

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## Polarized Low-Energy Positrons: A New Probe of Surface Magnetism

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A polarized beam of low-energy positrons has been used for the first time as a probe of surface magnetism. The polarization,  $P_e^-$ , of electrons captured at a Ni(110) surface to form positronium is measured. The temperature dependence of  $P_e^-$ , fitted by a power law, yields an exponent of  $\beta_1 = 0.7 \pm 0.1$ , in qualitative agreement with calculations of the critical exponent for the surface-layer magnetization. Rapid quenching of the ferromagnetic order is observed for submonolayer coverages of oxygen.

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The investigation of surface magnetism has recently become a topic of active theoretical and experimental interest. Calculations<sup>1-5</sup> of surface electronic properties of the ferromagnetic transition metals, performed with a thin slab geometry, predict band structure, charge and net spin densities, surface states, etc. The experimental

techniques now being developed to study surface magnetic properties include polarized photoemission<sup>6</sup> and field emission,<sup>7</sup> electron capture by ions in glancing collisions,<sup>8</sup> polarized low-energy electron scattering,<sup>9</sup> neutron reflection,<sup>10</sup> and ferromagnetic resonance absorption.<sup>11</sup> Experimental tests are limited by the requirement that

probes must be both spin sensitive and surface selective. Spin-polarized high-energy positrons from  $\beta$  decay that penetrate far below the surface have been used to study *bulk* magnetic properties.<sup>12</sup> Weakly penetrating low-energy positron beams have just recently been employed to study the interactions of positrons and positronium with surfaces,<sup>13,14</sup> positron diffraction,<sup>15</sup> surface phase transitions,<sup>14</sup> and adsorbed overlayers.<sup>14</sup> In addition, it was found<sup>16</sup> that such low-energy positron beams could be spin polarized with measured values of the polarization as high as 60%.<sup>17</sup>

In this Letter we report the first use of a spin-polarized low-energy positron beam to investigate surface magnetism. By measuring the asymmetry in formation of the triplet spin state of positronium (Ps) on Ni(110) when either the Ni magnetization or the positron-beam polarization is reversed, the spin polarization,  $P_{e^-}$ , of the captured electron can be deduced. Because of the screening of the valence electrons<sup>18</sup> Ps formation can only occur in the region of low electron density outside the surface<sup>18,19</sup> ( $\sim 2$  Å from the ion cores of the Ni surface atoms). Thus this new probe should be very sensitive to surface electronic and magnetic properties. In this work we demonstrate the feasibility of this new technique. By measuring the temperature dependence of the triplet Ps asymmetry we show, consistent with polarized electron scattering measurements,<sup>9</sup> that the surface magnetization in the range  $0.46 < T/T_c < 1.0$  differs markedly from the bulk magnetization. We also have found the measurements to be extremely sensitive to an adsorbed monolayer of oxygen.

The experiment is shown schematically in Fig. 1. Axially polarized positrons from a 20-mCi <sup>22</sup>Na source are thermalized in a special Be-Pt moderator that efficiently produces slow positrons ( $\sim 1.4 \times 10^4$ /sec) with a high degree of polarization,  $P_{e^+}$  (measured<sup>17</sup> to be  $0.5 \pm 0.03$ ). After electrostatic focusing into a beam and bending through  $90^\circ$  in a cylindrical mirror analyzer the transversely polarized positrons pass through an axial-magnetic-field spin rotator. The positron's spin is rotated by  $+90^\circ$  (up) or  $-90^\circ$  (down) before entering the UHV target chamber. The positron beam, at a selected energy from 300 to 1500 eV, is focused onto the surface of a  $2 \times 1 \times 0.1$  cm<sup>3</sup> Ni(110) single crystal which has been magnetically saturated parallel or antiparallel to the easy  $[1\bar{1}1]$  direction (either up or down) by a current pulse through the electromagnet. The penetration depth of the beam is less than the thermal

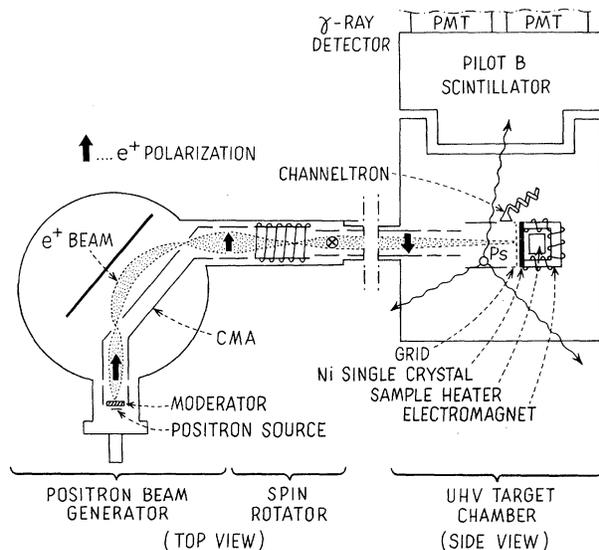


FIG. 1. The experimental arrangement. Details of the target chamber are rotated  $90^\circ$  for clarity.

diffusion length and thus virtually all positrons diffuse back to the surface where they may be trapped in a surface state,<sup>13</sup> capture an electron and leave the surface as Ps,<sup>19</sup> or be reemitted as a free positron and promptly returned to the surface by a grid biased  $+100$  V with respect to the target.

In order to distinguish decays due to triplet Ps an annihilation lifetime spectrum is acquired with use of a conventional time-to-amplitude converter-multichannel analyzer system. The time between the detection of secondary electrons ejected by the incident positron in a channel electron multiplier and the detection of the subsequent annihilation  $\gamma$  rays in a 12-in-diam Pilot B plastic scintillator is directly measured. This lifetime spectrum consists of a prompt peak due to free annihilation and singlet Ps decay, a flat background of random coincidences, and a single exponential component with a lifetime of  $\sim 110$  nsec due to decay of triplet Ps (the lifetime is reduced from 140 nsec by collisions with the lenses). We define  $R_T^+$  ( $R_T^-$ ) to be the number of triplet events in a time-delayed window between  $t=30$  and  $t=405$  nsec (after a background subtraction of 15% of the events) normalized to the total number of background-corrected events in the spectrum acquired with the magnetizing field up (down). The asymmetry  $A_T^\dagger$  in triplet Ps formation on reversing the magnetizing field is, for  $P_{e^+}$  up,

$$A_T^\dagger = (R_T^+ - R_T^-) / (R_T^+ + R_T^-). \quad (1)$$

To reduce systematics associated with residual fields from the magnet the spin rotator is then reversed to measure  $A_T^\dagger$  and  $A_T$  is taken to be  $(A_T^\dagger - A_T)/2$ .

The asymmetry in triplet positronium formation when a positron with polarization  $\vec{P}_{e^+}$  captures a polarized electron is  $\frac{1}{3}\vec{P}_{e^+} \cdot \vec{P}_{e^-}$ , where  $\vec{P}_{e^-}$  is the polarization of those electrons undergoing capture at the nickel surface. The experimentally detected asymmetry depends on detector geometry and is

$$A_T = \left[ \frac{2\epsilon(1) - \epsilon(0)}{2\epsilon(1) + \epsilon(0)} \right] \vec{P}_{e^+} \cdot \vec{P}_{e^-}, \quad (2)$$

where  $\epsilon(1)$  and  $\epsilon(0)$  are the  $\gamma$  detection efficiencies for annihilation from the  $m = \pm 1$  and  $m = 0$  states, respectively. With use of the calculated angular correlation functions,<sup>20</sup> the bracketed expression above is estimated to be  $0.49 \pm 0.04$ .

The Ni(110) sample is fastened between the pole faces of a C-shaped Armco iron electromagnet. Low-energy electron diffraction, Auger, and thermal desorption spectroscopy were used prior to the experiment in a separate surface-analysis chamber. Thermal desorption of CO was then used in the positron target chamber as a signature of a clean, well-characterized surface. The Ni(110) surface was cleaned by 2-keV Ar-ion sputtering and annealed at 850 °C. During a typical 3-h measurement of  $A_T$  the pressure in the target chamber was  $2 \times 10^{-10}$  Torr.

Values of  $A_T$  acquired in the initial phase of the experiment are shown in Table I. We note the following features: (a) Nonzero asymmetries that change sign<sup>21</sup> when the positron polarization is reversed are measured at magnetizing currents

TABLE I. Values of the Ps asymmetries,  $A_T^\dagger$  and  $A_T$ , measured on clean Ni(110) at various beam energies,  $E$ . The magnetizing current,  $I$ , is ramped up to the specified value and then down to zero prior to measuring asymmetries.

$E$ (eV)	$I$ (A)	$A_T^\dagger$ (%)	$A_T$ (%)
900	1	$-(0.23 \pm 0.20)$	$-(0.18 \pm 0.20)$
900	3	$-(0.23 \pm 0.18)$	$+(0.49 \pm 0.18)$
900	5	$-(0.22 \pm 0.21)$	$+(0.74 \pm 0.21)$
900	5	$-(0.08 \pm 0.08)^a$	
900	10	$-(0.06 \pm 0.18)$	$+(0.49 \pm 0.18)$
300	5	$-(0.63 \pm 0.06)$	$+(0.47 \pm 0.06)$
1500	5	$-(0.38 \pm 0.10)$	$+(0.36 \pm 0.10)$

<sup>a</sup> Spin rotator off.

of 3–10 A. At 1 A the effect disappears, consistent with magneto-optic Kerr-effect measurements that indicated that 2 A were required for saturation. (b) With the spin rotator off, and hence  $\vec{P}_{e^+} \perp \vec{P}_{e^-}$ ,  $A_T$  is zero as expected. (c) The beam energy and hence the relative contribution of thermal to nonthermal positrons diffusing back to the surface does not have a large effect on  $A_T$ . (d)  $A_T$  corresponds to the net capture of majority-spin electrons. With the inclusion of more recent<sup>21</sup> data  $P_{e^-} = (2.5 \pm 0.3)\%$  at room temperature.

As a check of the surface sensitivity of this new probe  $P_{e^-}$  was measured with an adsorbed overlayer of oxygen. After a room-temperature exposure of clean Ni(110) to 2 L [1 L (langmuir) =  $10^{-6}$  Torr sec] of  $O_2$  (0.5 monolayer)  $P_{e^-}$  decreased a factor of 5 to  $(0.5 \pm 0.2)\%$ . This quenching effect, which has also been observed in other surface experiments,<sup>9, 11</sup> clearly demonstrates that the captured electron does indeed come from the surface.

In order to show that this technique can be a quantitative probe of surface magnetism, we measured the temperature dependence of  $P_{e^-}$  from room temperature to the Ni Curie temperature,<sup>22</sup>  $T_c^B = 633$  K (Fig. 2). These data show the expected disappearance of  $P_{e^-}$  for  $T > T_c^B$ . Also shown in Fig. 2 are the results of a least-squares fitting of  $P_{e^-}(T)$  by the functional form

$$P_{e^-}(T) = P_{e^-}(0)(1 - T/T_c)^{\beta_1}. \quad (3)$$

The exponents shown represent extremum values of  $\beta_1$  for our fitting procedure. These data are not consistent with the bulk magnetization curve

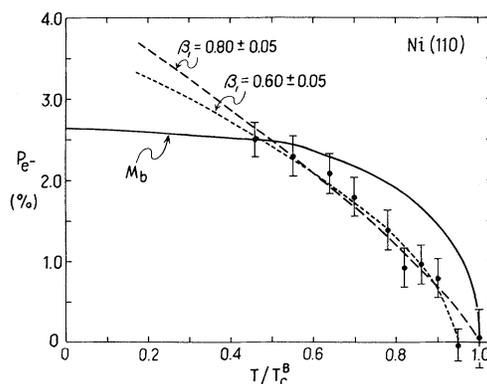


FIG. 2. Temperature dependence of  $P_{e^-}$ , the spin polarization of the captured electron. Dashed and dotted curves are fits of the data by Eq. (3) with  $T_c = T_c^B$  and with  $T_c$  unconstrained. The bulk magnetization,  $M_b$ , has been normalized to the datum point at  $T/T_c^B = 0.46$ .

( $\beta_b = 0.36$ ).<sup>22</sup> Our range for  $\beta_1$ ,  $0.7 \pm 0.1$ , is consistent with model calculations<sup>23,24</sup> (XY, Ising, Heisenberg) of the critical exponent of the surface-layer magnetization which range from approximately  $\frac{2}{3}$  to  $\frac{7}{8}$ . It is also in agreement with a polarized electron scattering experiment on Ni(110) which yielded  $\beta_1 = 0.77 \pm 0.02$ .<sup>24</sup> The temperature range over which  $P_{e^-}$  is measured is too broad for a rigorous determination of critical exponents. However, we do not expect distortion in  $P_{e^-}(T)$  due to the temperature dependence of the magnetic correlation length since Ps formation occurs only at the surface. In scattering experiments distortion in the surface-layer magnetization can result from the fact that the scattering process probes several atomic layers into the sample.<sup>9,23</sup>

Absolute comparison of our measured values of  $P_{e^-}$  with calculated surface-layer magnetic moments (which predict a 10% increase over the bulk Ni moment<sup>2,3</sup>) is complicated by the fact that the electron capture process does not equally sample all valence electronic states. The Ps work function is  $\varphi_{Ps} = \varphi_+ + \varphi_- - 6.8$  eV, where  $\varphi_+$  and  $\varphi_-$  are the positron and electron work functions (-1.5 and 5.2 eV, respectively), and 6.8 eV is the ground-state Ps binding energy. Thus Ps formation from thermal positrons is energetically allowed only for electrons within 3.1 eV of the Fermi level,  $E_F$ . Although the weighting function of the energetically allowed states is not yet known, our observation of a small majority-electron polarization on Ni(110) suggests a number of alternatives: (1) Since the electrons within 0.2 eV of  $E_F$  are predominantly of minority spin, the measurement of a net majority spin indicates that the capture process is only weakly energy dependent. (2) Both momentum and energy dependence may strongly modulate the capture process. Spin-polarized field emission, in which the tunneling probability is strongly energy and momentum dependent, also gives a small majority effect<sup>7</sup> on Ni(110). (3) The positron is not a weak perturbation to the ground state of the system, i.e., the screening cloud alters the initial-state spin distribution of the electrons.

In conclusion, we note that a number of future experiments are crucial to a more detailed understanding of the underlying capture process. These include the measurement of  $P_{e^-}$  on different crystallographic faces of Ni which could differentiate between alternatives 1 and 2 above. Measurement of  $P_{e^-}$  for thermally desorbed Ps,<sup>13</sup> where the captured electron does come from within a few

$kT$  of  $E_F$  (possibly on Fe or Co where  $T_c$  is high), should yield a large polarization<sup>25</sup> unless the screening cloud strongly alters the electron spin distribution. In addition, further measurements of the quenching of  $P_{e^-}$  for a variety of chemisorbed and physisorbed gases would be a rigorously quantitative test of surface magnetism theories.

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## Spin-Polarized Auger Spectroscopy from Magnetically Ordered Solids

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Auger electrons from magnetically ordered solids are spin polarized. As an example ferromagnetic Fe<sub>83</sub>B<sub>17</sub> glass was studied. The polarization of the Fe Auger electrons exhibits structures on the order of 20%, both parallel and opposite to the magnetization of the sample depending on the particular transition. Auger electrons from B are unpolarized. The technique opens new perspectives in the study of magnetic solids, particularly alloys and compounds.

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High-resolution Auger spectroscopy from solids yields rich information on electron and hole interactions.<sup>1</sup> LMM spectra from 3d metals contain the extent of delocalization of 3d holes in metals with unfilled d bands<sup>2</sup> and reveal the effective Coulomb interaction between two 3d holes at the same site.<sup>3</sup> Ferromagnetism, on the other hand, is probably the most important quality of the 3d transition metals and their compounds. In the present work we hope to open new perspectives in both the Auger spectroscopy on 3d transition metals as well as in the ferromagnetism of pure metals, compounds, and alloys, by studying the spin polarization of Auger electrons from a ferromagnetic solid. The information gained is manifold depending on the particular Auger transition. LVV (e.g.,  $L_3M_{45}M_{45}$ ) transitions provide a local probe of the spin-dependent density of valence states where special relevance has to be given to electron-electron correlation effects depending on the strength of the effective Coulomb interaction relative to the bandwidth. LMM transitions with no valence electrons involved, on the other hand, are spin polarized through the coupling of partly filled inner shells with the net spin of the magnetic 3d electrons,<sup>4</sup> and thus pro-

vide information on the local magnetization (i.e., average magnetic moment).

We wish to emphasize the element specificity of the Auger transition as well as its local character. We point out that spin-polarized Auger spectroscopy from solids therefore will be a powerful technique to study not only pure ferromagnetic metals but also magnetic alloys and compounds. Also ferrimagnetic systems will be of interest where sublattice specific information can be gained.

As a first example we present in this Letter spin-polarized Auger spectra from a ferromagnetic glass of composition Fe<sub>83</sub>B<sub>17</sub>. The sample was kindly provided to us by R. C. O'Handley at the Massachusetts Institute of Technology. In Figs. 1-3 we present intensity  $I(E_s)$  and spin polarization  $P(E_s)$  versus secondary-electron kinetic energy  $E_s$  in the vicinity of all the prominent Auger lines of iron and boron.

The spin polarization is defined as  $P(E_s) = [j^{\uparrow}(E_s) - j^{\downarrow}(E_s)] / [j^{\uparrow}(E_s) + j^{\downarrow}(E_s)]$ , with  $j^{\uparrow}(E_s)$  being the intensity of majority-spin electrons, i.e., electrons with magnetic moment parallel to the magnetization of the ferromagnetic sample. Polarization due to spin-orbit coupling in the