Spin-Polarized Metastable-Atom Deexcitation Spectroscopy: A New Probe of Surface Magnetism

M. Onellion, M. W. Hart, F. B. Dunning, and G. K. Walters

Department of Physics and The Rice Quantum Institute, Rice University, Houston, Texas 77251 (Received 17 November 1983)

It is demonstrated that spin dependences in the secondary-electron yields obtained when electron-spin-polarized $He(2^{3}S)$ atoms are deexcited at a magnetized surface provide a means by which to probe surface magnetism. This technique, which is particularly surface specific, has been used to investigate the dependence of Ni(110) surface magnetism on temperature and the presence of controlled adsorbates.

PACS numbers: 79.20.Rf, 73.20.-r, 75.30.Kz

In recent years there has been increasing interest in the study of surface magnetism and a variety of interesting phenomena have been observed by using polarized low-energy electron diffraction,¹ polarized photoemission,² electroncapture spectroscopy,³ and polarized low-energy positrons.⁴ Here we demonstrate a new method for probing surface magnetic properties based on spin dependences in the yields of secondary (Auger) electrons obtained when electron-spin-polarized $He(2^{3}S)$ atoms interact with a ferromagnetic surface. This technique, which is particularly surface specific because the incident atoms interact predominantly with the outermost atomic layer,⁵ has been used to investigate the dependence of Ni(110) surface magnetism on temperature and the presence of controlled adsorbates.

The apparatus, shown schematically in Fig. 1, has been described in detail elsewhere.⁶ A fraction of the atoms contained in a ground-state heli-

um beam are excited to the $2^{3}S$ level by coaxial electron impact. A weak (~2 G) transverse magnetic field is applied to define a quantization axis. Circularly polarized $1.08-\mu m \ 2^{3}S + 2^{3}P$ radiation incident along the magnetic field direction is used to optically pump the $2^{3}S$ atoms to increase the relative population in the $m_{j}(m_{s}) = +1$ or -1 magnetic sublevel. The resultant beam polarization P_{z} , ⁷ ~ 0.40, is measured by a Stern-Gerlach (SG) analyzer,⁶ and can be readily reversed, $P_{z} + -P_{z}$, by changing the sense of circular polarization of the pumping radiation. The SG system is also used to check that the He($2^{3}S$) beam contains no significant admixture of He($2^{1}S$) metastables, photons, or fast neutral atoms.⁶

A fraction of the electrons ejected when the $He(2^{3}S)$ beam strikes the target surface enter a retarding-grid energy analyzer, with energy resolution ~ 1-1.5 eV, and those with sufficient energy to surmount the retarding potential barrier



FIG. 1. Schematic diagram of the apparatus.

© 1984 The American Physical Society

are detected by a Channeltron. The transverse magnetic field restricts reliable electron energy distribution measurements to energies ≥ 3 eV.

The Ni(110) target crystal is mounted in the keeper position on a C -shaped electromagnet used to magnetize it along the easy [111] direction. Magneto-optic Kerr effect measurements⁸ showed that the crystal could be readily magnetized to saturation. The crystal is cleaned by argon-ion bombardment followed by annealing to $350 \,^{\circ}$ C. Auger analysis and low-energy electron diffraction demonstrated that this procedure provides a clean, well ordered surface.

Initial studies showed, as expected, that the secondary-electron yield from a clean, but unmagnetized, Ni(110) sample is independent of metastable-atom polarization. However, sample magnetization leads to a marked polarization dependence in the electron yield, which we characterize by an experimental asymmetry parameter

$$A = \frac{1}{|P_{\sigma}|} \frac{R_{+} - R_{-}}{R_{+} + R_{-}},$$

where R_+ and R_- are the Channeltron count rates for beam polarizations of $+P_z$ and $-P_z$, respectively. Measured values of A are shown in Fig. 2 as a function of the retarding bias applied in the energy analyzer. The corresponding secondary-electron energy distribution is shown in the inset.

The origin of the polarization dependence in the electron yield can be understood by reference to the energy-level diagram shown in Fig. 3. As a



FIG. 2. Polarization asymmetry A as a function of retarding potential for a clean, magnetized Ni(110) sample at ~ 130 °C. The inset shows the measured secondary-electron energy distribution.

 $He(2^{3}S)$ atom approaches a Ni(110) surface, it is first resonantly ionized by tunneling of the excited 2s electron into an unfilled level above the Fermi surface in the metal, as illustrated by the wavy arrow. The resulting He⁺ ion continues toward the surface where it is neutralized by a conduction electron from the metal, the released energy going to a second conduction electron which may, if the energy transferred is sufficiently large, escape from the metal. This two-electron process, termed Auger neutralization, leads to a distribution of ejected-electron energies, reflecting, approximately, a self-convolution of the local density of occupied electronic states at the surface.⁹ However, in this experiment, the electron spins in the incident $He(2^{3}S)$ atoms, and hence He⁺ ions, are polarized. If, as shown in Fig. 3, the electron in the ion is polarized spin "up," neutralization to the singlet ground state can occur only with a spin-down electron. Thus neutralization at a magnetized surface involves electrons in either the majority or minority spin bands, depending as to whether the initial $He(2^{3}S)$ polarization is parallel or antiparallel, respectively, to the surface magnetization.¹⁰ As illustrated schematically in the inset in Fig. 3, these bands are expected to be split in energy, as they are in the bulk, with the majority band lying entirely below the Fermi level. Since the highestenergy ejected electrons result from Auger neutralization processes in which the two electrons both originate near the Fermi level, where there are few majority-spin electrons, 10 A should be



FIG. 3. Energy level diagram illustrating the $He(2^{3})$ -Ni(110) surface interaction. Resonance ionization is indicated by the wavy arrows, Auger neutralization by corresponding pairs of straight arrows, and open arrows denote electron spins.

30 January 1984

largest at high energies, as is observed (see Fig. 2). As expected, the electron yield is greatest when the metastable atoms are polarized antiparallel to the bulk magnetization. This is also observed at lower energies, presumably because neutralization by electrons in the minority band will, on the average, release more energy than neutralization by electrons in the majority band, resulting in a higher secondary-electron escape probability.

The secondary-electron energy distribution is observed to be independent of temperature. Thus A should be directly proportional to the surface magnetization, which the data presented in Fig. 4 for several retarding potentials show to decrease approximately linearly with temperature. The extrapolated surface Curie temperature equals that of the bulk to within experimental uncertainty. This behavior is in reasonable agreement with studies by other spectroscopies,^{1,4} but is in marked contrast to bulk magnetization behavior.¹¹

As shown in Fig. 5, A decreases exponentially with surface exposure to CO, falling to ~0 at exposures ≥ 4 L, which constitutes saturation coverage.¹² Thus the CO-covered surface is nonmagnetic. As expected,¹³ exposure to CO also leads to changes in the electron energy distribu-



The present data indicate that measurements of spin dependences in secondary-electron ejection can be used to probe surface magnetism. The technique is attractive because it is particularly surface specific and because the incident metastable atoms do not modify the surface, either through sputtering or chemical reaction. With improved energy resolution and for magnetic materials with larger exchange splittings (e.g., Fe, magnetic glasses), we hope to extend this method to provide information on the spindependent local density of surface electronic states. We propose further to explore the potential of this technique for studies of materials such as chromium which, according to recent studies,^{3, 14} display surface ferromagnetism.

This work was supported in part by the Materials Sciences Section, Office of Basic Energy Sciences, U. S. Department of Energy, the Robert A. Welch Foundation, and the Donors of the Petroleum Research Fund administered by the American Chemical Society. One of us (M.O.) was the recipient of a Fannie and John Hertz Foundation Fellowship.



FIG. 4. Temperature dependence of the polarization asymmetry A for retarding potentials of, circles, -13V; triangles, -11 V; and squares, -9 V. The solid lines are the best straight-line fits to each data set. T_c indicates the bulk Curie temperature.



FIG. 5. Polarization asymmetry A as a function of exposure to CO at a crystal temperature of 50 °C for retarding potentials of, triangles, -11 V; and squares, -9 V. The inset shows the energy distribution for clean Ni(110) and following 1- and 4-L exposure to CO [1 L (langmuir) = 10^{-6} Torr sec].

¹R. J. Celotta, D. T. Pierce, G.-C. Wang, S. D. Bader, and G. P. Felcher, Phys. Rev. Lett. <u>43</u>, 728 (1979); S. Alvarado, M. Campagna, F. Ciccacci, and H. Hopster, J. Appl. Phys. <u>53</u>, 7920 (1982).

²H. Hopster, R. Raue, G. Güntherodt, E. Kisker, R. Clauberg, and M. Campagna, Phys. Rev. Lett. <u>51</u>, 829 (1983).

³C. Rau, J. Magn. Magn. Mater. 30, 141 (1982).

⁴D. W. Gidley, A. R. Köymen, and T. W. Capehart, Phys. Rev. Lett. <u>49</u>, 1779 (1982).

⁵H. D. Hagstrum, Phys. Rev. <u>96</u>, 336 (1954); J. A. Appelbaum and D. R. Hamann, Phys. Rev. B <u>12</u>, 5590 (1975).

⁶T. W. Riddle, M. Onellion, F. B. Dunning, and G. K. Walters, Rev. Sci. Instrum. <u>52</u>, 797 (1981), and <u>53</u>, 257 (1982).

 ${}^{7}P_{z} = (n_{+} - n_{-})/(n_{+} + n_{0} + n_{-})$, where n_{+} , n_{0} , and n_{-} are the fluxes of atoms with $m_{j}(=m_{s}) = +1$, 0, and -1, respectively.

⁸P. N. Argyses, Phys. Rev. <u>97</u>, 334 (1955).

⁹For a detailed discussion of secondary-electron ejection by metastable atoms, see H. D. Hagstrum, in *Electron and Ion Spectroscopy of Solids*, edited by L. Fiermans, J. Vennik, and W. Dekeyser (Plenum, New York, 1978), pp. 273-323.

¹⁰The 3*d* electrons dominate the secondary-electron spectrum, especially at higher energies where *A* is largest. However, the nonmagnetic 4*s* electrons, which we disregard here, also presumably contribute to the observed spectrum and dilute the spin asymmetry.

¹¹J. D. Cohen and T. R. Carver, Phys. Rev. B <u>15</u>, 5350 (1977).

¹²W. Erley and H. Wagner, J. Chem. Phys. <u>72</u>, 2207 (1980).

¹³See, for example, F. Bozso, J. T. Yates, Jr., J. Arias, H. Metiu, and R. M. Martin, J. Chem. Phys.

78, 4256 (1983), for the effects of CO on Ni(111). ¹⁴F. Meier, D. Pescia, and T. Schriber, Phys. Rev. Lett. 48, 645 (1982).