

Spin dependence in He(2^3S) metastable-atom deexcitation at magnetized Fe(110) and O/Fe(110) surfaces

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Spin-labeling techniques, specifically the use of electron-spin-polarized He(2^3S) metastable atoms coupled with analysis of the number and spin of the ejected electrons, are used to investigate the dynamics of metastable-atom-surface interactions and the properties of (magnetized) Fe(110) and O/Fe(110) surfaces. The data show that the dominant He(2^3S) metastable-atom deexcitation mechanism at such surfaces is resonance ionization followed by Auger neutralization, and that the ejected-electron polarization reflects the iron conduction-band polarization. The present results, when interpreted using the theory of Penn and Apell, also indicate that the magnetization in the vacuum above a clean Fe(110) surface at distances ($\sim 3\text{--}5 \text{ \AA}$) where Auger neutralization occurs is negative but changes sign upon exposure to oxygen. Several possible explanations for this sign reversal are discussed. The measurements establish spin-polarized metastable-atom deexcitation spectroscopy as an extraordinarily sensitive probe of the surface magnetic environment.

Earlier work in this laboratory using a magnetized Ni(110) surface has demonstrated that spin-polarized metastable-atom deexcitation spectroscopy (SPMDS) can provide information on surface magnetic properties.¹ In SPMDS a beam of electron-spin-polarized He(2^3S) metastable atoms is directed at the target surface. The polarization of the electrons ejected from the surface as a result of metastable-atom deexcitation is measured, together with (for a magnetized surface) any spin dependence in the total ejected-electron signal. In the present paper we report the results of a SPMDS study of an Fe(110) surface, and of the same surface following exposure to oxygen. The oxygen-iron system is of particular interest in view of recent theoretical and experimental findings that there is substantial exchange splitting in the oxygen-adsorbate-induced bands for both the O/Fe(100) (Refs. 2-4) and O/Fe(110) (Ref. 5) systems. The present data provide insights into the dynamics of metastable-atom deexcitation at surfaces and on the magnetization in the near-surface vacuum.

The present apparatus is shown schematically in Fig. 1 and is similar to that used in earlier studies.^{1,6} Briefly, a fraction of the atoms contained in a ground-state helium-atom beam are collisionally excited to the $2^{1,3}S$ levels by a coaxial electron beam. The 2^1S atoms are removed from the beam by illuminating it with $2.05\text{-}\mu\text{m}$ radiation from a helium discharge which excites $2^1S \rightarrow 2^1P \rightarrow 1^1S$ transitions. A weak ($\sim 0.5 \text{ G}$) magnetic field is applied perpendicular to the beam to preserve a well-defined quantization axis. Circularly polarized $1.08\text{-}\mu\text{m}$ $2^3S \rightarrow 2^3P$ resonance radiation from a high-power rf-excited helium lamp is incident along the magnetic field direction and is used to optically pump the 2^3S

atoms to increase the relative populations in the $M_S = +1$ or -1 magnetic sublevels. The resultant beam polarization is defined as

$$P_{\text{He}} = \frac{F_+ - F_-}{F_+ + F_0 + F_-}, \quad (1)$$

where F_+ , F_0 , and F_- are the fluxes of He(2^3S) atoms with $M_S = +1$, 0 , and -1 , respectively. The beam polarization, $P_{\text{He}} \sim 0.4$, is measured by a Stern-Gerlach analyzer and can be simply reversed ($P_{\text{He}} \rightarrow -P_{\text{He}}$) by changing the sense of circular polarization of the optical pumping radiation.

The Fe(110) target was a film of thickness $\sim 3000 \text{ \AA}$ grown epitaxially (in a separate apparatus) on a GaAs substrate. Its surface was cleaned by Ne⁺-ion bombardment, followed by annealing at 200°C until a good low-energy electron diffraction (LEED) pattern was observed. The low anneal temperature is necessary to avoid As diffusion into the iron film and limited the Ne⁺-ion energies that could be used in sputtering to $\lesssim 400 \text{ eV}$. Consequently, it was not possible to completely remove all surface contaminants. Auger spectroscopy showed that following cleaning there remained $\sim 9\%$ carbon and $\sim 4\%$ oxygen on the surface. The sample was pulse magnetized to saturation along the [110] easy axis and all data were acquired in the remanent state. *In situ* magneto-optical Kerr-effect measurements yielded square hysteresis loops and verified that full single-domain magnetization was preserved in remanence.

Oxygen adsorption on room-temperature Fe(110) is a complex process that has been investigated using a number of techniques.^{7,8} For surface coverages up to

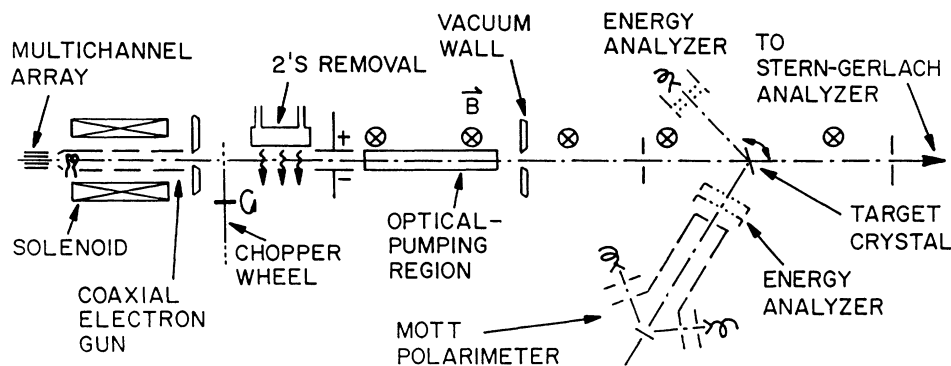


FIG. 1. Schematic diagram of the apparatus.

$\Theta_O \sim 0.4$ chemisorption occurs and a sequence of different LEED patterns [$c(2 \times 2)$, $c(3 \times 1)$, and split $c(3 \times 1)$] is observed as the oxygen exposure, and thus coverage, is increased. At higher coverages oxygen begins to penetrate the surface as a precursor to oxide formation. Although there are discrepancies in the earlier work as to the relationship between surface coverage and oxygen exposure, the present exposures (up to 16 L, where 1 L = 1 langmuir = 10^{-6} Torr sec) should be sufficient to obtain a coverage of at least $\Theta_O \sim 0.4$.

The energy distribution of the electrons ejected from the target surface is measured using a simple retarding-grid energy analyzer, and those electrons with sufficient energy to overcome the retarding-potential barrier are detected by a channeltron. The number of electrons with energies in some particular interval is determined by switching the potential applied to the retarding grid between the appropriate limits and observing the resultant *change* in the detected electron signal. The energy analyzer is also used to investigate spin dependences in the total number of electrons ejected with energies greater than the cutoff determined by the potential V applied to the retarding grid. Such spin dependences are characterized by an asymmetry parameter $A(V)$ defined as

$$A(V) = \frac{1}{|P_{\text{He}}|} \frac{I_+ - I_-}{I_+ + I_-}, \quad (2)$$

where I_+ and I_- are the ejected-electron currents observed with the incident beam polarized parallel and antiparallel, respectively, to the majority-spin direction in the (magnetized) target. The observed asymmetries were, however, quite small and this precluded energy-resolved asymmetry measurements using the same differential technique as employed to determine ejected-electron energy distributions.

A second independent experimental parameter, the polarization of the ejected electrons, is also measured. This is accomplished using a compact Mott polarimeter equipped with a retarding-potential-energy analyzer.⁹ The average polarization of those ejected electrons with energies greater than the cutoff set by the retarding potential V applied in the energy analyzer is determined by measuring the asymmetry in the count rates of electrons

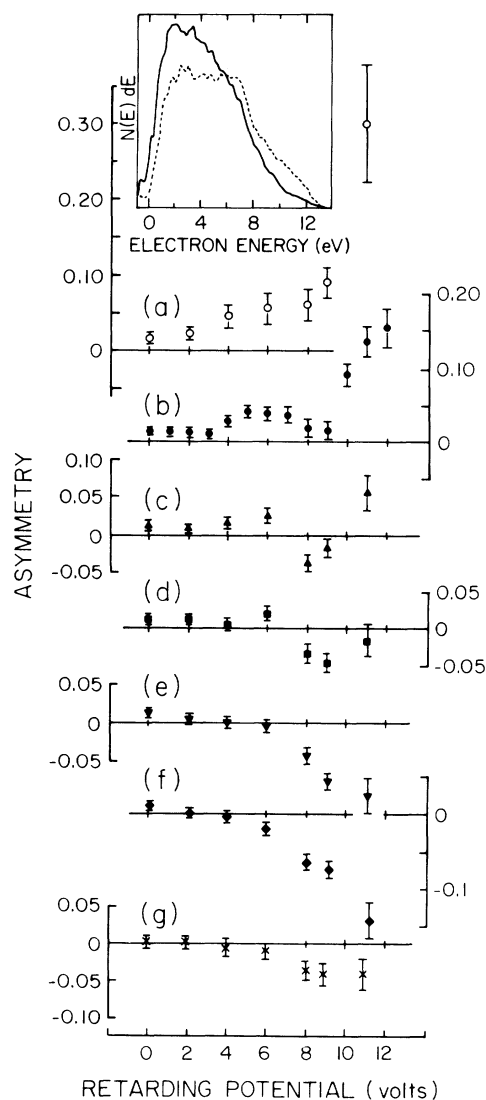


FIG. 2. Values of the asymmetry $A(V)$ for (a) an atomically clean Fe(100) surface (obtained by extrapolation, see text); (b) a sputtered and annealed Fe(110) surface; and (c), (d), (e), (f), and (g) an Fe(110) surface following exposure to 1, 2, 4, 8, and 16 L of oxygen, respectively. The inset shows the ejected electron-energy distributions for a clean Fe(110) surface (---) and following an 8-L exposure to oxygen (—).

quasielastically scattered (at 20 keV) through $\pm 120^\circ$ at a gold target. The electron polarization, which depends both on the helium-atom polarization and the retarding potential, is defined by

$$P_e(P_{\text{He}}, V) = \frac{I_P - I_A}{I_P + I_A}, \quad (3)$$

where I_P and I_A are the currents of electrons ejected with spins parallel and antiparallel, respectively, to the majority-spin direction in the target.

Values of the asymmetry $A(V)$ measured for the cleanest Fe(110) surface obtained (to be referred to simply as the "clean" surface) and following oxygen exposures of 1, 2, 4, 8, and 16 L are shown in Fig. 2. The ejected-electron energy distributions for the clean surface, and following 8-L exposure to oxygen, are shown in the inset. Figure 2 also includes values of the asymmetry expected for an atomically clean Fe(110) surface ($\Theta_O=0$) obtained by linear extrapolation of the asymmetries measured at different oxygen coverages as inferred from Auger analysis. (Tests revealed that the presence of small amounts of carbon on the surface had a negligible effect on the measured asymmetries.)

The measured polarizations $P_e(P_{\text{He}}, V)$ of electrons ejected from the clean Fe(110) surface, and following exposure to 8 L of oxygen, are presented in Table I for several values of the energy cutoff, i.e., the retarding potential V . Table I includes data obtained using an unpolarized incident beam, and an incident beam polarized both parallel and antiparallel to the majority-spin direction in the (magnetized) sample. Since the polarizations of both the incident He(2^3S) atoms and ejected electrons are defined relative to the target majority-spin direction, the measured values of P_e should remain unchanged upon reversal of the magnetization of the target ($M \uparrow \rightarrow M \downarrow$), as is observed.

The asymmetries $A(V)$ for the clean Fe(110) surface are positive, and have the same sign as observed in our earlier studies of Ni(110).¹ The asymmetries are, however, very sensitive to the presence of adsorbed oxygen,

especially at the higher retarding potentials where the sign of the asymmetries reverses for exposures $\gtrsim 2$ L. The asymmetry magnitudes peak at about 8 L exposure and tend toward zero for higher exposures, perhaps corresponding to the onset of oxide formation.^{7,8} In contrast, the ejected-electron energy distribution is relatively insensitive to oxygen exposure. Further, the polarization of electrons ejected by an unpolarized incident beam is uniformly positive and is essentially unchanged by oxygen exposure. The data in Table I also reveal a significant spin correlation favoring the ejection of electrons with the same spin orientation as the incident metastable atoms. This spin correlation, which amounts to $\sim 20\%$, i.e., the change in polarization of the ejected electrons is $\sim 20\%$ that of the incident atoms, is similar to that observed previously at paramagnetic surfaces.⁶

The experimental results will be discussed with reference to both the theory of SPMDS recently developed by Penn and Apell¹⁰ (PA) and to the conventional models of metastable atom deexcitation at surfaces first proposed by Hagstrum.¹¹ If a He(2^3S) atom is incident on a clean, high-work-function metal surface, such as Fe(110) ($\phi \cong 5.1$ eV), it is first assumed to undergo resonance ionization (RI) in which the excited $2s$ electron tunnels into an unfilled level above the Fermi surface. The resulting He⁺ ion continues toward the surface, where it undergoes Auger neutralization (AN), in which a conduction electron from the metal tunnels into the $1s$ hole, the energy released being communicated to a second (Auger) conduction electron which may escape from the metal. The energy available to the escaping electron depends on the energy of the He⁺ $1s$ hole and this decreases as the ion approaches the surface due to the He⁺ image potential. Thus, the farther from the surface that the AN event occurs, the greater the energy (on the average) of the escaping electron. Alternately, if the incident He(2^3S) atoms can get close to the surface without undergoing RI, direct Auger deexcitation (AD) may occur. This is the dominant deexcitation mechanism (a) for surfaces of work function sufficiently low that RI cannot occur because the resonance states of appropriate energy in the

TABLE I. Measured spin polarization $P_e(V)$ of electrons ejected by both unpolarized He(2^3S) atoms and by He(2^3S) atoms polarized parallel and antiparallel to the majority-spin direction in the target. Data for the clean Fe(110) surface after reversing the target magnetization ($M \uparrow \rightarrow M \downarrow$) are included. The majority-spin direction in the target is taken to be the direction of positive P_{He} and $P_e(V)$. The uncertainty in each measured value of P_e is $\sim \pm 0.03$.

| | V | $M \uparrow$ | | | $M \downarrow$ | | |
|------------------------------|-----|-----------------|----------|----------|-----------------|----------|----------|
| | | P_{He} | $P_e(V)$ | $P_e(V)$ | P_{He} | $P_e(V)$ | $P_e(V)$ |
| | | -0.40 | 0 | +0.40 | -0.40 | 0 | +0.40 |
| (a) Clean Fe(110) | 0 | 0.07 | 0.12 | 0.21 | 0.004 | 0.10 | 0.21 |
| | 3 | 0.02 | 0.08 | 0.18 | 0.04 | 0.09 | 0.15 |
| | 6 | -0.01 | 0.07 | 0.14 | 0.02 | 0.04 | 0.19 |
| | 9 | -0.06 | 0.10 | 0.22 | 0.02 | 0.11 | 0.21 |
| (b) O/Fe(110) (8-L exposure) | 0 | 0.04 | 0.10 | 0.15 | | | |
| | 3 | -0.03 | 0.07 | 0.14 | | | |
| | 6 | -0.02 | 0.08 | 0.15 | | | |
| | 9 | -0.03 | 0.09 | 0.21 | | | |

metal lie below the Fermi level and hence are already occupied, or (b) when the RI step is suppressed by the presence of an adsorbate. Under either of these circumstances, an incident He(2^3S) atom undergoes AD, the He(2^3S) $1s$ hole being filled by an electron from the metal surface [case (a)] or from the adsorbate [case (b), also termed surface Penning ionization], with the simultaneous ejection of the He $2s$ electron.

In the present work a polarized He(2^3S) atom beam is employed. In AD it is the $2s$ atomic electron that is liberated, which will therefore have a polarization equal to that of the incident beam. In contrast, for RI + AN, the ejected electron originates in the surface. Thus any detected ejected-electron polarization must result either from differences in the densities of minority- and majority-spin states at the surface or from a correlation in spin orientation of the ejected and neutralizing electrons involved in the AN process. [The polarization of the neutralizing electron must be opposite to that of the He $^+$ $1s$ electron, i.e., parent He(2^3S) atom, because the helium ground state is a spin singlet.]

The ejected-electron polarizations resulting from the deexcitation of an unpolarized He(2^3S) beam at both a clean Fe(110) surface, and following oxygen exposure, are positive, i.e., the ejected-electron polarization is parallel to the majority-spin direction and reflects the conduction-band polarization. This requires that, on average, the ejected electrons originate deeply enough within the target surface that their polarizations are not significantly affected by the presence of an oxygen adlayer. Photoemission¹² and electron-bombardment studies¹³ of magnetized iron surfaces also have shown that the ejected-electron polarization mirrors the band polarization, but the measured polarizations are much greater than observed in the present work. The present data are, however, in reasonable agreement with the results of Kirschner *et al.*, who studied the polarization of electrons ejected (by potential ejection) when 1-keV He $^+$ or Ar $^+$ ions are neutralized at a magnetized Fe(110) surface.¹⁴ They attribute the generally low polarizations of the ejected electrons to a reduction in the band polarization in the near-surface region, where the ejected electrons originate, because of the spill out of negatively polarized s - p electrons above the surface. Alternately, matrix-element effects favoring Auger ejection from the s - p band could account for the low measured polarization.

Two possible interpretations can be advanced to explain the spin correlation evident in Table I. If it is asserted that incident metastable atoms are deexcited exclusively by RI + AN, the observed spin correlation would imply that the electrons involved in the AN process tend to have antiparallel spins. This would require that AN favor singlet two-hole final states in the target as has been reported for certain Auger processes originating on core holes in ferromagnets.¹⁵ An alternate explanation has also recently been examined, namely that RI + AN and AD occur in parallel, in which event the observed spin correlation can be attributed to the AD component without assuming any significant spin correlation in the AN process.¹⁶ Calculations show that a He(2^3S)

atom is strongly perturbed as it approaches a surface as a consequence of image-charge effects. In particular, because of mixing of the $2s$ and $2p_z$ orbitals, the probability density associated with the excited electron becomes markedly asymmetric and peaked toward vacuum. As a result, the electron-tunneling rate is reduced. This could allow a fraction of the atoms in an incident beam to survive passage to atom-surface separations sufficiently small that AD becomes important. However, the observed spin correlation is small ($\sim 20\%$) and thus, even if AD is occurring, RI + AN must still be the dominant deexcitation process at both the clean and oxygen-covered surfaces.

It is puzzling that the presence of the oxygen adlayer with the attendant modification in the near-surface electronic and magnetic environment results in so little change in the ejected electron polarizations, especially given a spin correlation that favors the ejection of an electron with the same spin orientation as the He(2^3S) atom that produces it. From the definition [Eq. (2)] of the asymmetry, $A(V) > 0$ implies that a positively polarized incident He(2^3S) atom [i.e., a He(2^3S) atom with electron spins parallel to the majority-spin direction in the target] has a greater probability of ejecting an electron with kinetic energy greater than the cutoff determined by V than does a negatively polarized He(2^3S) atom. Conversely, for $A(V) < 0$, negatively polarized 2^3S atoms have the greater probability of ejecting electrons. Thus, even for an unpolarized incident He(2^3S) beam, a majority of the electrons detected for any retarding voltage V for which $A(V) > 0$ (< 0) will be produced by the positively (negatively) polarized beam component. For $A > 0$, spin correlation therefore favors the ejection of positively polarized electrons, and for $A < 0$ it favors negatively polarized electrons. This suggests that for an unpolarized He(2^3S) beam the spin polarizations from the clean surface ($A > 0$) should be greater than those from the oxygenated surface ($A < 0$), because for $A > 0$ both the band polarization and spin correlation favor negative polarization. Assuming the band polarization to be no greater for the oxygenated surface than for the clean surface, spin-correlation effects should result in a measured spin polarization for the clean surface that is significantly larger than for the oxygenated surface. While the experimental uncertainties in P_e are too large to preclude that possibility, the data of Table I do not suggest such a difference.

The asymmetries shown in Fig. 2 can be discussed in terms of the theory of SPMDS recently developed by Penn and Apell (PA),¹⁰ which is based on the RI + AN deexcitation mechanism and suggests that the measured asymmetries result as a consequence of a nonzero magnetization in the region well outside the surface where AN occurs. As noted previously, because of image-charge effects, the energy available to the ejected electron increases monotonically with the perpendicular distance z from the target surface at which AN occurs. PA point out that the AN rate at some distance z is proportional to the number density of conduction electrons at z that are available to fill the He $^+$ $1s$ hole. For incident He(2^3S) atoms, and hence He $^+$ ions, with positive (negative) po-

larizations [i.e., spins parallel (antiparallel) to the majority-spin direction in the magnetized target] neutralization can only occur with minority (majority) conduction electrons (the helium ground state is a spin singlet). Thus the average distance at which AN occurs, and hence the average energy available to the Auger electron, will depend on the spin orientation of the incident He(2^3S) atoms because the majority- and minority-spin densities in the vacuum are in general unequal for a ferromagnetic target.

Using this model, PA have shown that the sign of the asymmetry parameter $A(V)$ will be positive (negative) if the conduction-electron density in the vacuum outside the target surface is predominantly of minority-spin (majority-spin) type. Their analysis of our earlier data for Ni(110) yields a magnetization of $\sim -20\%$ at the Fermi energy and 4.5 Å from the surface.¹¹ The negative magnetization in the vacuum is consistent with band calculations of Wimmer *et al.*, who show that the nickel $s-p$ electrons, which, through $s-d$ hybridization, are polarized oppositely from the d electrons that dominate the total magnetic moment in the bulk, spill out into the vacuum region and are dominant beyond ~ 2.5 Å.¹⁷ Negative magnetization in the vacuum above Ni(110) has also been reported by Rau, based on electron-capture experiments.¹⁸

The present observation that $A(V) > 0$ for a clean Fe(110) surface requires, on the basis of the PA theory, that minority electrons be dominant at distances of $\sim 3-5$ Å outside the surface where AN occurs. As for nickel, it is known that the $s-p$ electrons in iron are negatively magnetized,¹⁹ and the calculations of Noffke²⁰ indeed suggest a negative polarization well outside the Fe(110) surface. However, Onishi *et al.* calculate that for Fe(100) the magnetization in the near-surface vacuum ($\lesssim 2.5$ Å) is positive.²¹ Further theoretical calculations of spin densities as a function of distance from Fe(110) and O/Fe(110) surfaces out to about 5 Å would provide a quantitative test of the PA theory.

The present suggestion that the vacuum magnetization above an Fe(110) surface is negative is in disagreement with Winter *et al.*, who concluded from their experiments on the resonant capture of electrons by fast ions incident at grazing angle on a magnetized Fe(110) target that the magnetization in the vacuum is in fact positive at distances comparable to those where AN occurs.²² However, electron-capture distances and mechanisms in such experiments are poorly understood at present, posing interpretational difficulties. The large ion momentum parallel to the target surface severely distorts the Fermi sea in the ion reference frame and capture is predominantly from states which have total momentum in the plane of the surface.^{23,24} In SPMDS the neutralizing electrons in the AN event are predominantly those with total momentum *normal* to the surface, i.e., those with greatest extension into the vacuum.¹¹

The measured asymmetries are extremely sensitive to the presence of oxygen on the surface, reversing sign and becoming strongly negative for exposures above about 4 L. According to the PA theory, this requires that the vacuum magnetization at distances where AN occurs

change from negative to positive upon oxygen adsorption. A number of factors might contribute to reversal of the vacuum magnetization. Firstly, the total conduction-electron density at AN distances is very small, and it is reasonable to presume that their involvement in the bonding of chemisorbed oxygen to the Fe(110) surface could significantly alter both the total density and the balance between majority- and minority-spin electrons in the vacuum, especially in view of the fact that the oxygen adsorbate bands are known to be exchange split as a result of strong ferromagnetic coupling to the substrate. Indeed, there is both experimental and theoretical evidence suggesting the participation of iron $s-p$ electrons in the oxygen chemisorption bonds.^{2,4,25}

The spin-polarized photoemission measurements of Schönhense *et al.*⁵ on the O/Fe(110) surface provide further insight into the interpretation of the asymmetry data. Their data reveal a broad spin-split O $2p$ band centered about 6 eV below the Fermi level. Examination of the asymmetry data for low oxygen coverages (Fig. 2) reveals that the oxygen-induced asymmetry changes originate with a significant dip about 6 eV below the high-energy limit of the ejected-electron energy distribution. The high-energy limit corresponds to AN events in which both neutralizing and ejected electrons originate at the Fermi level. The asymmetry dip would be accounted for if He⁺ ions incident upon an adsorbed oxygen site were neutralized by predominantly positively polarized electrons near the Fermi level, with the ejected electrons coming from the O $2p$ band. The negative asymmetry in this case would be reinforced as a consequence of the exchange splitting of the $2p$ band, because the electron-spin-correlation effect discussed earlier will favor ejection of the higher-energy minority electrons when the incident beam is polarized antiparallel to the target majority-spin direction.

In summary, we conclude that the dominant mechanism for He(2^3S) metastable-atom deexcitation at both clean and oxygen-exposed Fe(110) surfaces is RI+AN. Positive asymmetries are observed for clean Fe(110) which, according to the PA theory of SPMDS, implies a negative magnetization in the vacuum outside the surface at distances where AN occurs ($\sim 3-5$ Å). The asymmetry, however, is very sensitive to oxygen exposure, and reverses sign, suggesting that the near-surface vacuum magnetization becomes positive. Several possibilities that might account for the reversal are suggested. In contrast to the measured asymmetries, the spin polarization of electrons ejected from clean and oxygen-exposed Fe(110) are nearly identical and reflect the positive band polarization of the target surface. The measurements establish SPMDS as an extraordinarily sensitive probe of the surface magnetic environment and the pronounced changes that can result upon exposure to ambient gases.

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