

PLEED STUDIES OF THE TEMPERATURE DEPENDENCE OF THE SURFACE
VS BULK MAGNETIZATION OF Ni(110) *

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The temperature dependence of the bulk and surface magnetization of Ni(110) is derived using the molecular field theory. These results are then used in a dynamical polarized low-energy-electron-diffraction (PLEED) calculation to yield the temperature dependent polarization of the diffracted intensities, which is compared with recent experimental data for $0.6 T_{\text{Curie}} \leq T \leq T_{\text{Curie}}$. The importance of multiple scatterings and the usefulness of PLEED for surface magnetic structural determination are clearly demonstrated.

Submitted to Solid State Communications

* Work supported by the Department of Energy, contract DE-AC03-76SF00515.

It is well known in atomic physics that the elastic exchange scattering of a spin polarized electron beam with hydrogen atom produces a polarization effect. This polarization effect is caused by the difference of the singlet ($e\uparrow + H\downarrow$) and triplet ($e\uparrow + H\uparrow$) scattering cross sections derived from using the antisymmetric and symmetric spin state wave functions for the two-electron systems according to the Pauli exclusion principle. Similarly, in the case of elastic polarized low-energy electron diffraction (PLEED) from ferromagnetic surfaces, in which all the surface spins are aligned in one direction via the application of an external magnetic field, the exchange scattering is determined by the interaction of the incident electron spin (\vec{S}_1) with the surface spin (\vec{S}_2) components which are parallel (\uparrow) or antiparallel (\downarrow) to \vec{S}_1 . This produces a difference, $I^\uparrow - I^\downarrow$, in the diffracted intensities. The polarization S is defined as $S = (I^\uparrow - I^\downarrow)/(I^\uparrow + I^\downarrow)$. However, in practice it is impossible to calculate exactly the scattering amplitude by using antisymmetrized wavefunctions for an infinite electron system. Instead, various approximations were made to handle this problem. Among those the local spin density functional formalism has proved most useful. In this approach, one replaces the nonlocal exchange correlation potential between the incident electron and the surface by a spin-dependent local potential $H_{int}^{\uparrow,\downarrow}$ and the problem of antisymmetrization is then reduced to a spin dependent potential scattering. In this work we analyze the polarization of the diffracted electrons from a clean Ni(110) surface and its temperature dependence near the Curie point.

Figure 1 shows the scattering geometry in which \vec{S}_1 is made to lie within the scattering plane to eliminate the spin-orbit effect. Ni(110) is magnetized along the easy magnetization axis $[1\bar{1}1]$, which is parallel to the surface.

$$H_{int}^{\uparrow,\downarrow}(\mathbf{r}) = \sum_j V(\vec{r}-\vec{R}_j) + \sum_j V_{xc}^{\uparrow,\downarrow}(\vec{r}-\vec{R}_j) + iV_{oi} \quad (1)$$

where V is the Coulomb potential, $V_{xc}^{\uparrow,\downarrow}$ is the spin-dependent local exchange-correlation potential, V_{oi} is the imaginary part of the scattering potential which simulates the damping of the incident wave field. The spin

dependence of V_{0i} is found to be negligible for Ni.¹ \vec{R}_j is the position of each Ni atom. For $T=0$, we used the best available band structural potential for $\text{ReH}_{\text{int}}^{\uparrow,\downarrow}$ obtained by Moruzzi, Janak and Williams.² The authors used the muffin-tin approximation and an exchange-correlation-potential suggested by von Barth and Hedin.³ It can be represented as a shifted and rescaled spin dependent Slater potential, i.e.,

$$V_{\text{xc}}^{\uparrow,\downarrow}(T=0) = A(\rho) \left(\frac{2\rho^{\uparrow,\downarrow}}{\rho} \right)^{1/3} + B(\rho) \quad (2)$$

where $\rho = \rho^{\uparrow} + \rho^{\downarrow}$ is the total electron density and A,B are potentials used in the spinless case by Hedin and Lundqvist.⁴ The above V_{xc} should strictly be used only for bound electrons, however it has also generally been used for PLEED calculations. Since the energy splitting between the "spin up" and "spin down" bands of Ni is a few tenths of an eV, which is much larger than the thermal energy, electronic excitations from "spin up" bands (which have lower energy) to "spin down" bands (which have higher energy) is very unlikely. This implies that the spin magnetic moment per Ni atom at $T > 0$ is essentially unchanged. On the other hand, the spin rotational energy is small and comparable to the thermal energy and provides the main mechanism for the disappearance of magnetic ordering as $T \rightarrow T_C$, where T_C is the Ni bulk Curie temperature (~ 632 K). We then have to modify (2) to take into account the temperature effect. By linearizing (2), we have

$$V_{\text{xc}}^{\uparrow,\downarrow}(T>0) \sim A(\rho) + B(\rho) + 1/3A(\rho) \left(\frac{\rho_s}{\rho} \right)^{1/3} \langle \vec{\mu}_1 \cdot \vec{\mu}_2 \rangle_T \quad (3)$$

where $\rho_s = \rho^{\uparrow} - \rho^{\downarrow}$, and $\vec{\mu}_1$ and $\vec{\mu}_2$ are unit vectors. $\vec{\mu}_1 \cdot \vec{\mu}_2 = 1$ or -1 , if the incident electron spin vector $\vec{\mu}_1$ is parallel or antiparallel to the surface spin vector $\vec{\mu}_2$ and T denotes a thermal average. Taking $\vec{\mu}_1$ to be along the z axis, then what we need to know is $\langle \vec{\mu}_2, z \rangle_T$. The calculation of this quantity for the surface case remains unsolved within the itinerant band theory. Instead, we adopt a simple Heisenberg local spin model and use the molecular field theory (MFT). The physical assumption of MFT is to consider only one magnetic atom and replace its interaction with the remainder of the crystal by an effective field. Also we assume that spins on the same layer have the same thermal averaged value.

Then the layer-dependent magnetization $m_n = \langle S_z \rangle_T / S = \langle \vec{\mu}_{2,z} \rangle_T$ of the n-th layer can be obtained as

$$m_n = B_S \left[\frac{S}{kT} \sum_{n'} J(n, n') m_{n'} \right] \quad (4)$$

where $B_S [\dots]$ is the Brillouin function and $J(n, n')$ is the layer-layer exchange coupling constant. S is the size of the Ni magnetic moment. $\sum_{n'}$ is included to only the nearest neighbors in this work. For Ni(110) (F.C.C.), (4) becomes a set of coupled equations which can be solved numerically.

$$\begin{aligned} m_1 &= B_S \left[\frac{S}{4t(S+1)} (2jm_1 + 4m_2 + m_3) \right] \\ m_2 &= B_S \left[\frac{S}{4t(S+1)} (4jm_1 + 2m_2 + 4m_3 + m_4) \right] \\ m_3 &= B_S \left[\frac{S}{4t(S+1)} (m_1 + 4m_2 + 2m_3 + 4m_4 + m_5) \right] \\ &\vdots \\ m_n &= B_S \left[\frac{S}{4t(S+1)} (m_{n-2} + m_{n+2} + 4(m_{n-1} + m_{n+1}) + 2m_n) \right] \end{aligned} \quad (5)$$

where $t = T/T_c$ and $j = J(1,1)/J$; otherwise $J(n, n') = J$ for any n, n' . These results are shown in Fig. 2. $j=1$ has been used for results shown in Figs. 4-6.

The magnetic correlation length ξ becomes infinite at $t=1$ and decreases as T decreases below T_c . The surface magnetization is smaller than the bulk value, but no dead layers are found. We can see that it is useful to use PLEED to study surface magnetization when the electron mean-free path is less than ξ . When t is very close to 1, (5) can be approximated by the analytic expression

$$m_n \sim m_{\text{bulk}} \cdot \tanh(n\phi + \delta) \quad (6)$$

where

$$m_{\text{bulk}} = \frac{\sqrt{1-t}}{C_s}$$

$$\phi = \sqrt{\frac{6}{5}} (1-t)$$

$$\delta = \frac{4(j-1)\sqrt{3(1-t)}}{1+4(1-j)}$$

Here $C_S^2 = 3/5[S(S+1)+1/2](S+1)^{-2}$. It is then clear that $m_{\text{bulk}} \propto (1-t)$ and $m_1 \propto (t-1)$ as $t \rightarrow 1$.

This behavior can be found in Fig. 3, where the T dependence of m_{bulk} and m_1 , etc., is clearly demonstrated.

The results of (5) are then used in (3) to obtain $V_{\text{xc}}^{\uparrow,\downarrow}(T>0)$, i.e.,

$$V_{\text{xc}}^{\uparrow,\downarrow}(T>0) \sim \frac{1}{2} \left[V_{\text{xc}}^{\uparrow}(T=0) - V_{\text{xc}}^{\downarrow}(T=0) \right] \pm m_n \times \Delta V_{\text{xc}} \quad (7)$$

where $\Delta V_{\text{xc}} = V_{\text{xc}}^{\uparrow}(T=0) - V_{\text{xc}}^{\downarrow}(T=0)$. "+" is for " \uparrow " and "-" is for " \downarrow ".

For the PLEED calculations, we used (7) to compute the phase shifts for e-atom scattering at each T for each Ni layer. We then compute diffraction intensities with the Renormalized-Forward-Scattering method⁶ with an appropriately modified LEED program.⁷ An inner potential of 11 eV and an energy dependent V_{oi} taken from elsewhere⁸ are used, as well as a 5% contraction of the top layer spacing, as determined by LEED studies.⁸

Fig. 4 shows the calculated I - E (intensity vs energy) and S - E (polarization vs energy) curves at $T = 0.6 T_c$ of Ni(110) for incident polar angle of $\theta = 12^\circ$ and incident azimuth along $[1\bar{1}1]$. Both the case with bulk magnetization in all layers and the case where each layer has our calculated magnetizations are shown. The S - E curves show pronounced structural differences between these two cases at $E = 100 - 150$ eV. We investigate in particular the S vs t dependence for $0.6 \leq t \leq 1$ at $E = 125, 124, 21$ and 20 eV shown in Figs. 5 and 6 at $\theta = 12^\circ$ and 60° . For the first three cases, the bulk S(t) curve always shows larger curvature at $t \rightarrow 1$ than the surface S(t) curve, although the latter includes actually the averaged magnetizations of many different layers. This trend correlates well with the $m_{\text{bulk}}(t) \propto (1-t)$ and $m_1(t) \propto (t-1)$ predictions in (6). This is also the trend predicted by using the LEED kinematic approximation⁹ and is in fair agreement with experimental data taken at $E = 125$ eV,¹⁰ although the theoretical S(t) values are somewhat lower than the experimental values. This discrepancy may be due to the following facts: (1) the experimental results have not taken the contact potential difference into account, (2) the experimental angle of incidence is not precisely 12° since the experimental curves (Fig. 3) of ref. 10 are not symmetrical, (3) the experimental S(t) values are

not normalized and, (4) approximations have been made in the theory. The trend obtained at $E = 21$ eV is however completely reversed and indicates the breakdown of the kinematic approximation. The $S(t)$ curve is therefore highly energy-dependent and nonkinematical features can easily occur near the sharp variation region of the S vs E curve.

These results support the use of PLEED for surface magnetic structural determinations.

Acknowledgements — The author wishes to thank Drs. R. E. Kirby, E. L. Garwin and M. A. Van Hove for reading the manuscript and Dr. D. T. Pierce for sending an experimental curve before its publication.

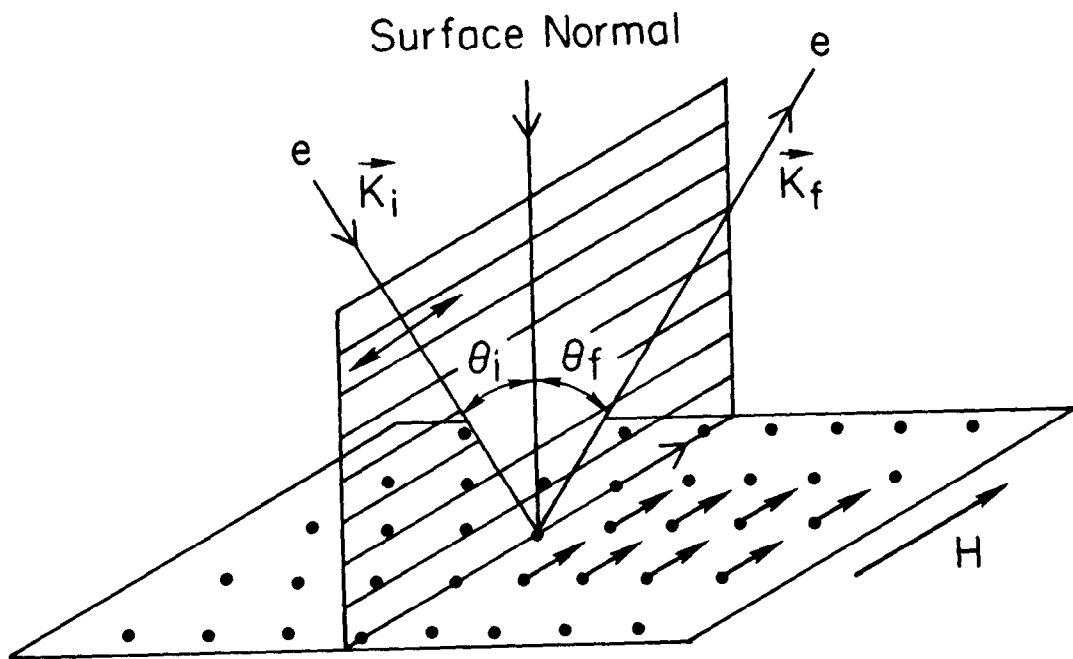
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* Work supported by the Department of Energy, contract DE-AC03-76SF00515.

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Figure Captions

1. Scattering geometry for the PLEED experiment.
2. Layer dependent magnetization $m_n(t)$ as a function of layer number n .
3. The layer dependent $m_n(t)$ as a function of t .
4. I-E and S-E curves for $\theta = 12^\circ$, $\phi = 35.26^\circ$ from 15 to 150 eV.
5. S vs t curve at $E = 124$ and 21 eV. Experimental data are taken from Reference 10 at $E = 125$ eV.
6. S vs t curve at $E = 125$ and 20 eV, $\theta = 12^\circ$, 60° and $\phi = 35.26^\circ$. λ_{expt} is the same line drawn through the experimental data points as shown in Fig. 5.



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Fig. 1

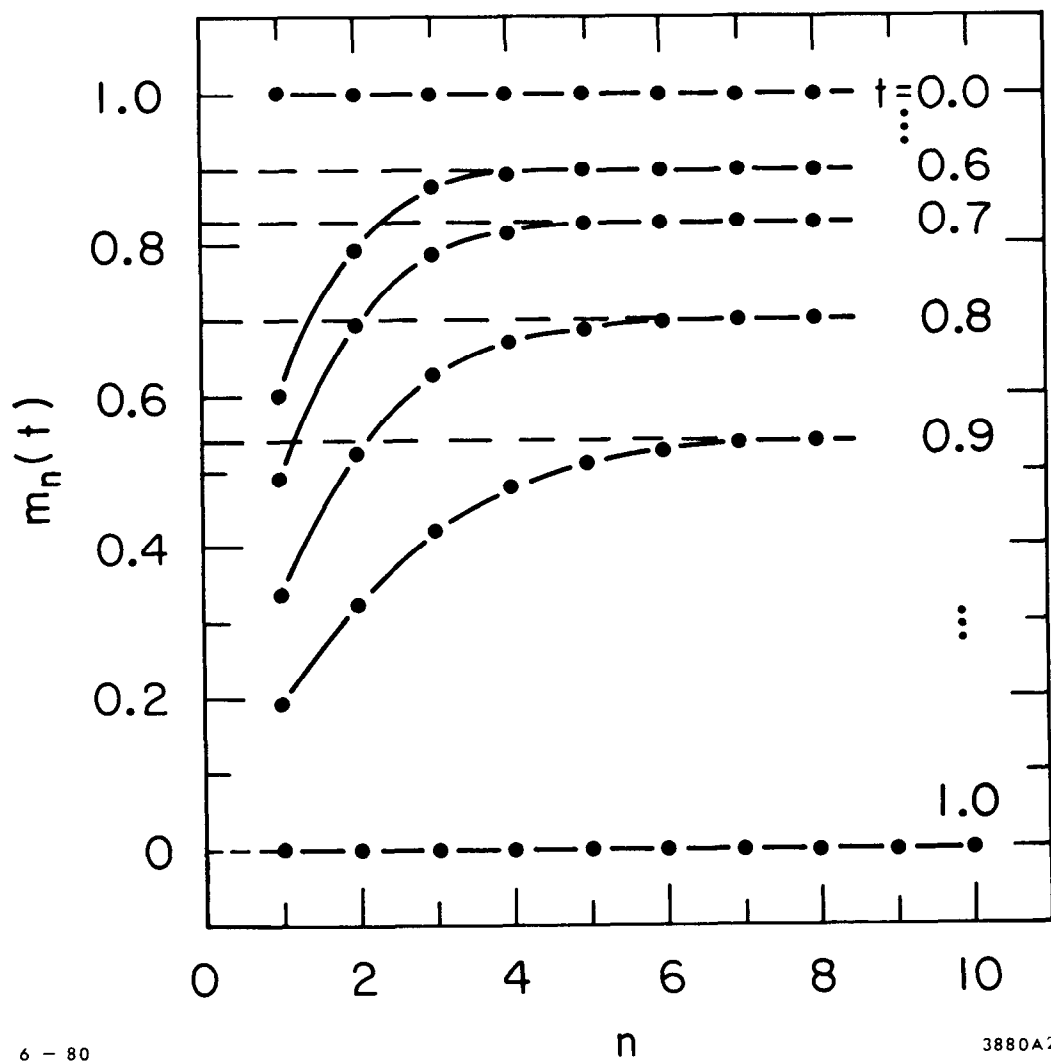


Fig. 2

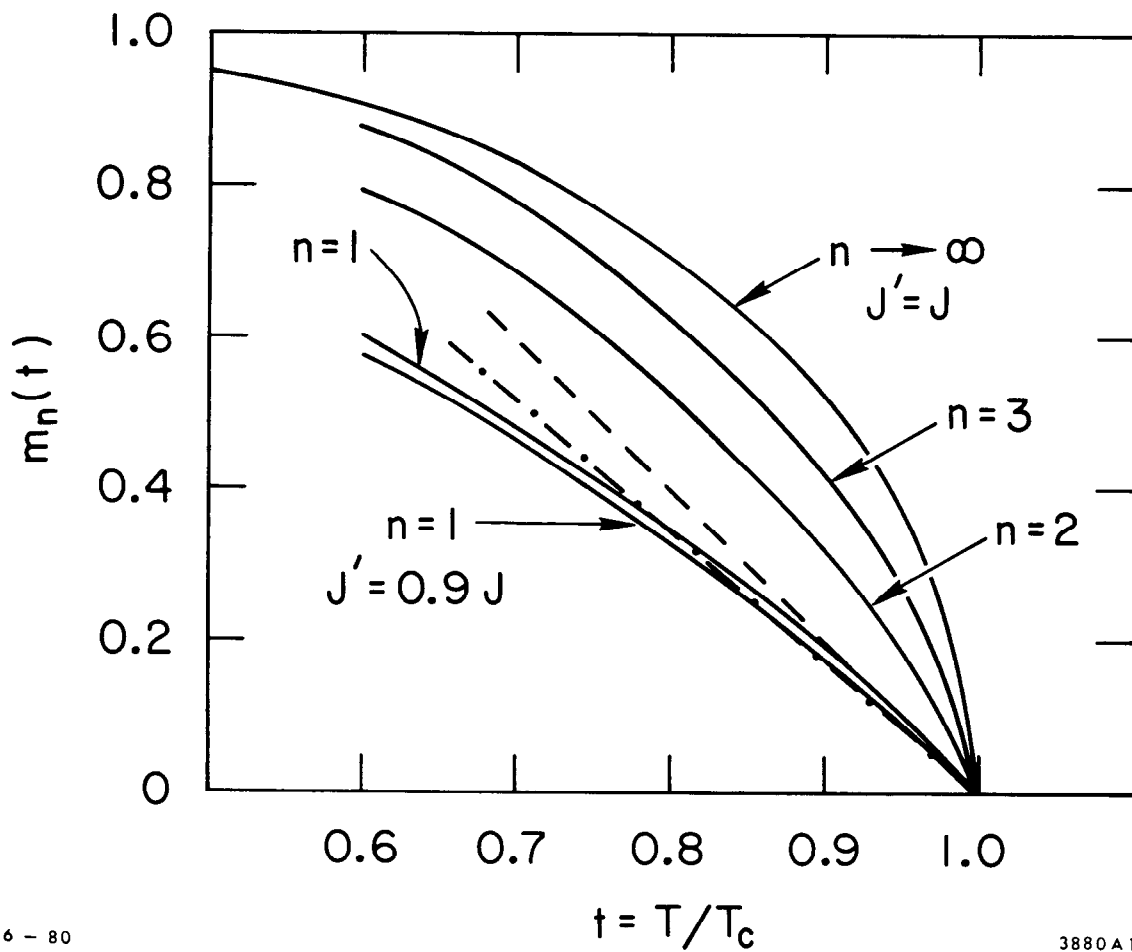


Fig. 3

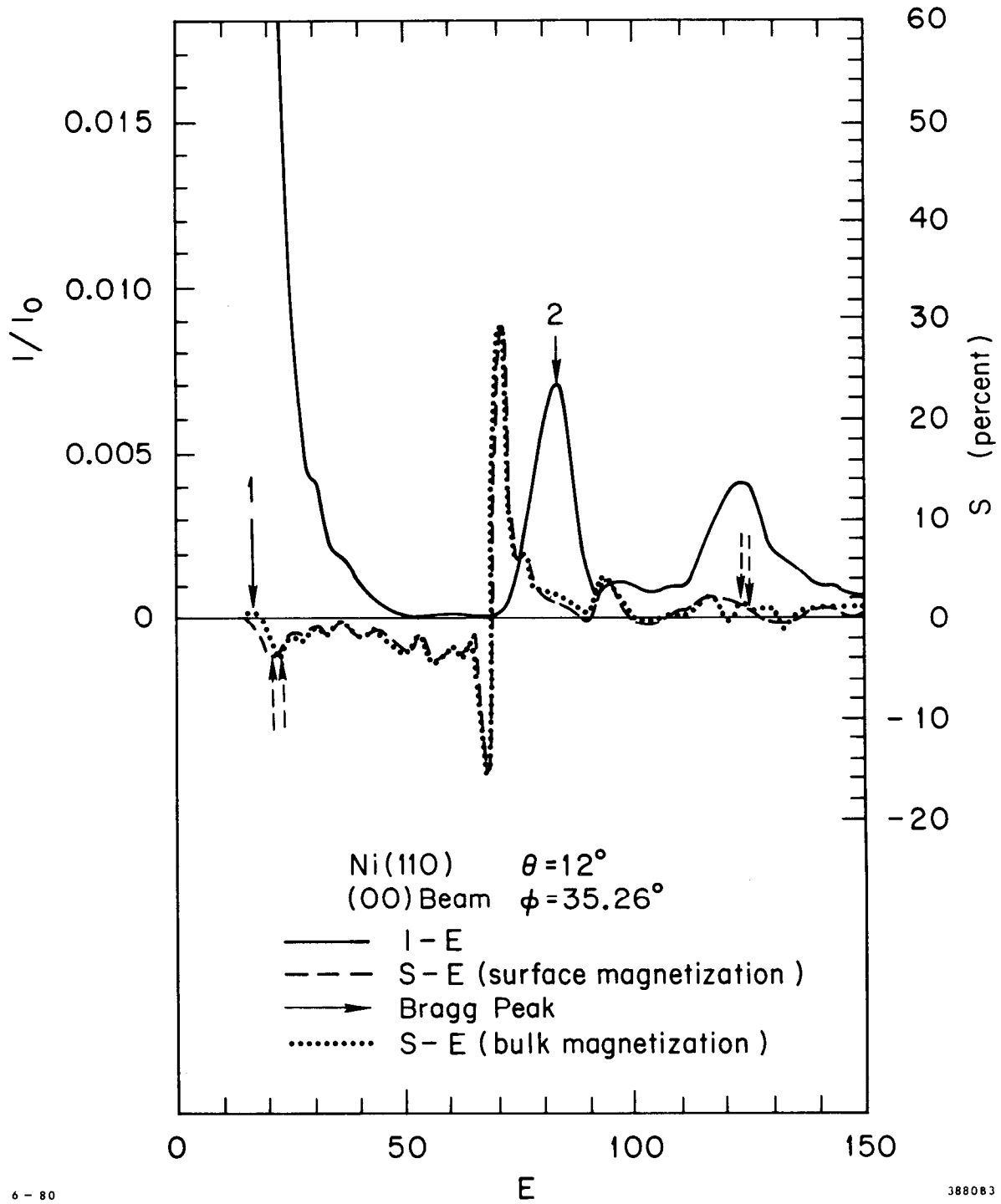


Fig. 4

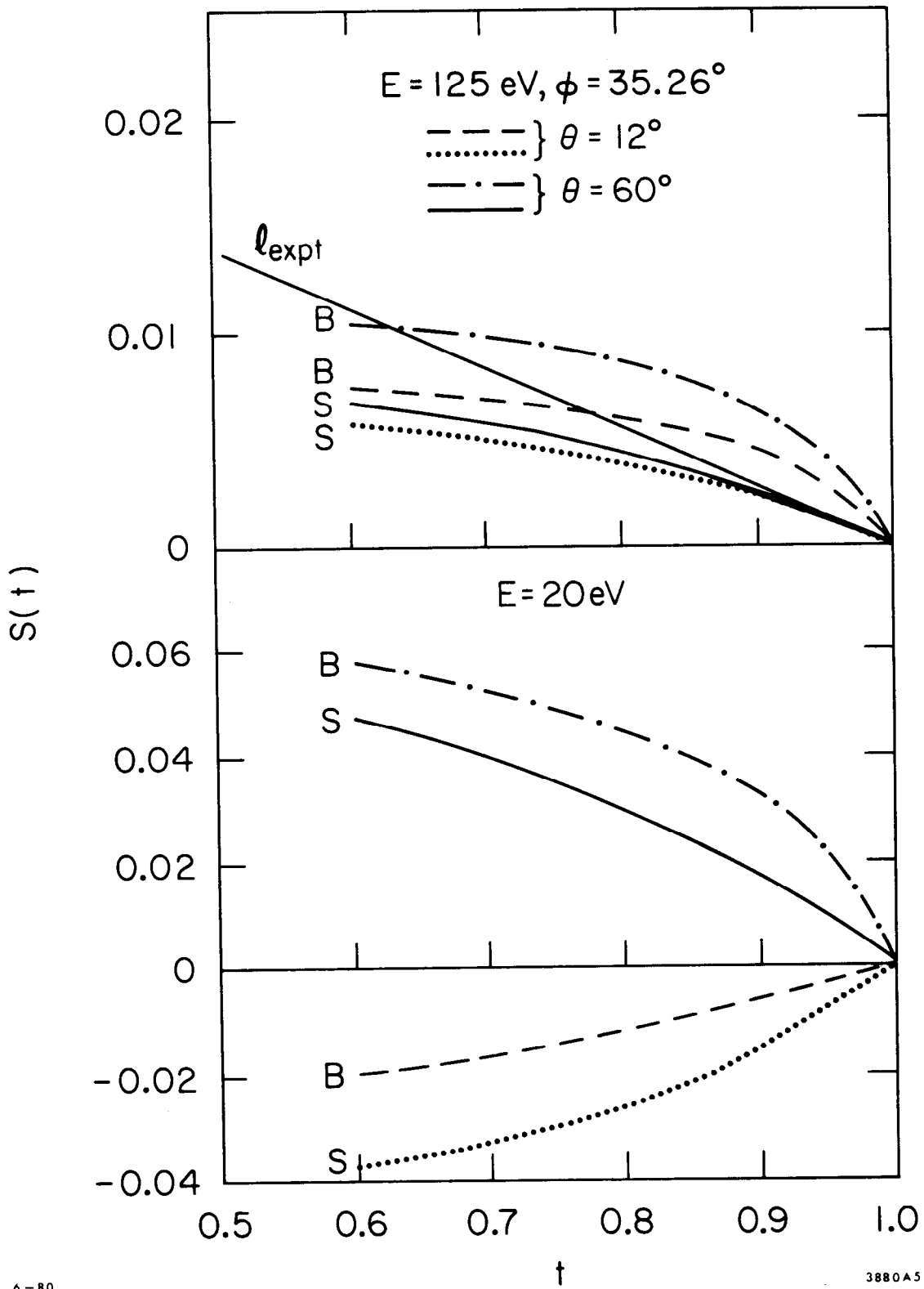


Fig. 5

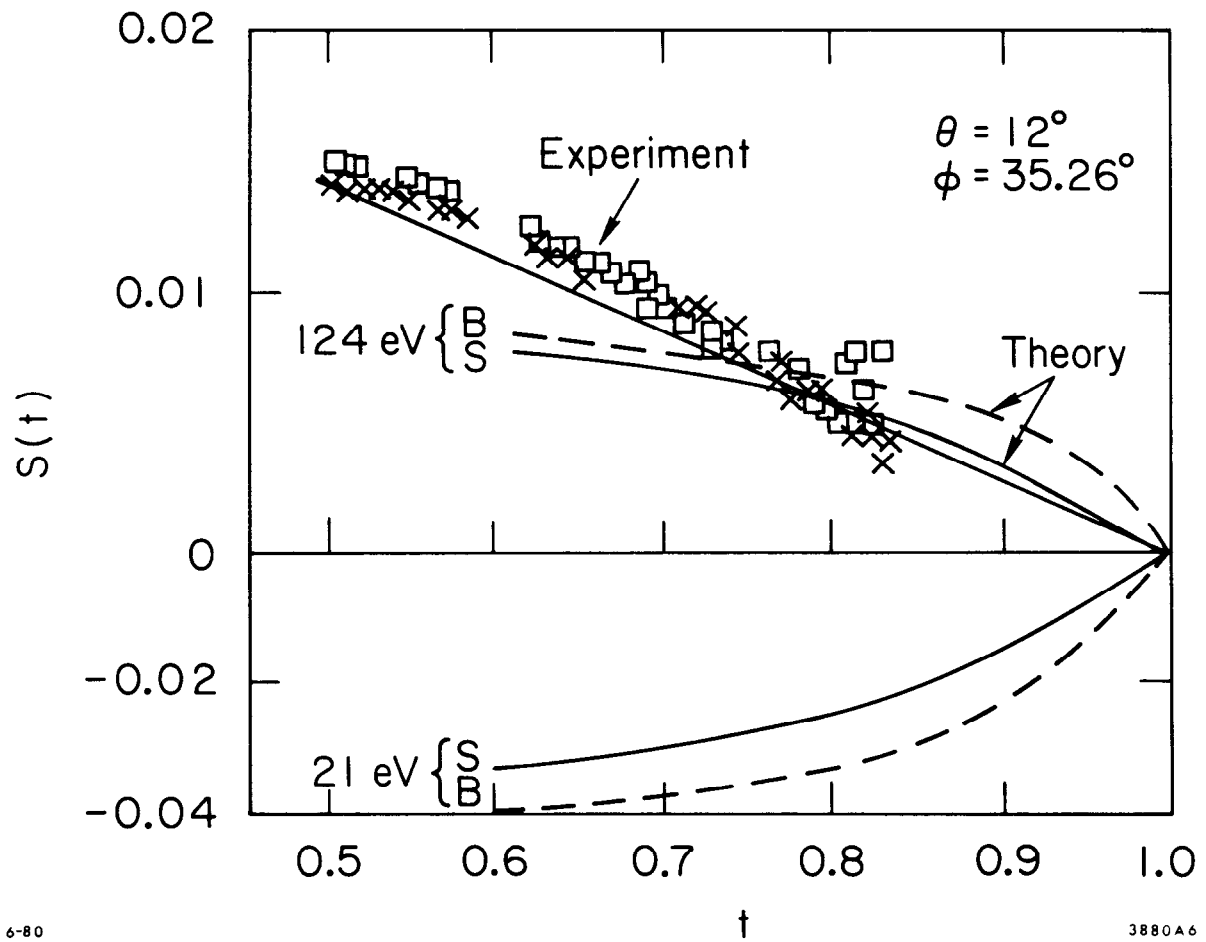


Fig. 6