ring group at the Physical Sciences Laboratory.

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Band-Structure Calculation of the Electron Spin Polarization in Field Emission from Ferromagnetic Nickel*

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A detailed $3d$ band structure of ferromagnetic Ni is incorporated into a calculation of the electron spin polarization in field emission from the (100) crystal plane. In agreement with a recent experiment of Gleich, Regenfus, and Sizmann, the calculated electron spin polarization is found to -4%, with the preferred spin direction parallel to the magnetization of the crystal.

Several recent measurements of electron spin polarization (ESP) in photoemission[1,2] and field emission[3] from Ni have produced controversy concerning the validity of the Stoner-Wolfarth-Slater (SWS) itinerant band model of ferromagnetism.[1-5] The following Letter describes the first application of a detailed SWS band structure in a rigorous calculation of the ESP in field emission from the (100) plane of ferromagnetic Ni. The results agree in sign and magnitude with ESP measurements of Gleich, Regenfus, and Sizman[3] for field emission around the [100] crystallographic direction of Ni.

The essential features of the model employed in the calculation are as follows: The emission or "z" direction coincides with the [001] direction of a semi-infinite fcc crystal of Ni. The emission surface at $z=0$ is $\frac{1}{2}a$ away from the last atom layer present, where $a=3.5$ Å is the interatomic spacing for fcc Ni. It is assumed the emission plane exhibits the two-dimensional translational symmetry of a (100) plane of the infinite lattice, and hence the condition for specular reflection and transmission is satisfied. The surface potential is represented by a triangular barrier given by $V(z)=-eFz$, $z>0$, with F denoting the applied electric field. The SWS itinerant band model is invoked to account for the ferromagnetic properties of Ni. Specifically, the unhybridized $3d$ and $4s$-p conduction bands are divided into identical spin bands, majority ($\uparrow$) and minority ($\downarrow$), separated by the exchange energies $\Delta E_{\mathrm{exch}}{}^{(d)}=0.03$ Ry and $\Delta E_{\mathrm{exch}}{}^{(s)}=0$. The $4s$-p spin bands are approximated by parabolic free-electron bands originating at Γ_0. The $3d$ bands are described by a tight-binding calcula-

tion originally formulated by Fletcher[6] but modified to agree with the minority-spin bands of Connolly.[7] The $3d$ energy bands, the eigenfunctions, and the constant-energy surfaces are derived exactly from the tight-binding secular determinant and used without approximation in the field-emission calculation. Of particular interest is the $3d$ Fermi surface which consists of three sheets, a highly irregular closed surface and two hole sheets.[8] In agreement with other energy-band calculations, the present results indicate that the majority-spin $3d$ bands do not contribute to the Fermi surface.

The field-emission current is treated as separately arising from the unhybridized $3d$ and $4s$-p spin bands. If $J_T^{\uparrow(\downarrow)}$ denotes the total field-emitted current of either spin, then $J_T^{\uparrow(\downarrow)} = J_{3d}^{\uparrow(\downarrow)} + J_{4s\text{-}p}^{\uparrow(\downarrow)}$. The resulting spin polarization P of the emitted beam is

$$P = \frac{J_T^{\uparrow} - J_T^{\downarrow}}{J_T^{\uparrow} + J_T^{\downarrow}} = \frac{J_{3d}^{\uparrow} - J_{3d}^{\downarrow}}{J_{3d}^{\uparrow} - J_{3d}^{\downarrow} + 2J_{4s\text{-}p}^{\uparrow}}, \quad (1)$$

assuming that $J_{4s\text{-}p}^{\uparrow} = J_{4s\text{-}p}^{\downarrow}$ since $\Delta E_{\text{exch}}^{(s)} = 0$.

Consider the calculation of $J_{3d}^{\uparrow(\downarrow)}$ due to tight-binding $3d$ Bloch states. It has been traditional to compute the field-emitted current as the product of a "supply function" $n(\vec{k})$, defined as the electron flux incident on the surface barrier, and $D(\vec{k})$, the probability that an electron in state $\vec{k}$ will be transmitted through the barrier. The total current is then $J = e\sum_{\vec{k}} n(\vec{k})D(\vec{k})$, where the sum is over all states $\vec{k}$.[9] However a recent critical study[8] of tunneling from three-dimensional Bloch states has shown that this expression is only completely valid for free electrons. First, $n(\vec{k})$ is usually derived with the aid of macroscopic kinetic arguments which are only completely rigorous for the noninteracting electron gas. Of more importance is the realization that in tunneling from a real three-dimensional crystal, the usual concept of a transmission coefficient $D(\vec{k})$ is ambiguous.[10] The ambiguity arises because there exists a nonvanishing interference current between "incident" and "reflected" Bloch states (i.e., where the group velocities are $\partial E_n/\partial\hbar k_z > 0$ and $\partial E_n/\partial\hbar k_z < 0$, respectively). Therefore, we calculate directly $\vec{J}^e$, the field-emitted current at the detector:

$$\vec{J}^e = (\hbar/2mi) \times [\psi_{II}^*(\vec{r})\nabla_{\vec{r}}\psi_{II}(\vec{r}) - \psi_{II}(\vec{r})\nabla_{\vec{r}}\psi_{II}^*(\vec{r})]_{\text{det}}, \quad (2)$$

where $\psi_{II}(\vec{r})$ is the wave function in the half-space $z > 0$. For the assumed one-dimensional surface potential, ψ_{II} is separable and of the form,

$$\psi_{II}(\vec{r}) = \sum_{\vec{G}_\parallel} C(\vec{G}_\parallel) e^{i(\vec{k}_\parallel + \vec{G}_\parallel)\cdot\vec{\rho}} f_{\vec{G}_\parallel}(z), \quad z > 0. \quad (3)$$

$\vec{\rho} = x\hat{x} + y\hat{y}$, and $\vec{G}_\parallel$ are the projections of the reciprocal lattice vectors on the (001) plane; $f_{\vec{G}_\parallel}(z)$ is a solution to the one-dimensional Schrödinger equation including the applied field. The constant coefficients $C(\vec{G}_\parallel)$ are obtained by matching ψ_{II} (and its z derivative) across the entire $z = 0$ emission surface to the solution ψ_I inside the metal:

$$\psi_I = \psi_{n\vec{k}} + \sum_{n'\vec{k}'} A(n'\vec{k}')\psi_{n'\vec{k}'}, \quad z < 0, \quad (4)$$

where $E_n(\vec{k}) = E_{n'}(\vec{k}')$, $\vec{k}_\parallel = \vec{k}_\parallel$ (condition of specular reflection), and $\partial E_n(\vec{k})/\partial\hbar k_z > 0$ and $\partial E_{n'}(\vec{k}')/\partial\hbar k_z < 0$. This makes the $C(\vec{G}_\parallel)$, and ultimately $\vec{J}^e$, explicit functions of the indices $(n, \vec{k})$. The details of this matching procedure have been presented elsewhere[11]; we state only that it entails the evaluation of the $G_\parallel$th Fourier components of the projections of the $3d$ eigenfunctions on the $z = 0$ plane.

We define $\bar{j}_z^{\uparrow(\downarrow)}(n, \vec{k})$ as the emitted current density per Bloch state $(n, \vec{k})$, averaged over a planar detector of area S parallel to the emission surface[12]:

$$\bar{j}_z^{\uparrow(\downarrow)}(n, \vec{k}) = S^{-1}\int_S \vec{J}^e \cdot \hat{z}\, da.$$

The resulting expression for $\bar{j}_z(n, \vec{k})$ is very complicated, but has the following functional form:

$$\bar{j}_z^{\uparrow(\downarrow)}(n, \vec{k}) = P(n, \vec{k}) \exp\left[-\frac{4}{3}\frac{\sqrt{\alpha}}{F}\left(\frac{k_\parallel^2}{\alpha} - E\right)^{3/2}\right], \quad (5)$$

where $E = E_n^{\uparrow(\downarrow)}(\vec{k})$ (in eV) is a negative energy measured from zero in vacuum, F is in V/Å, $\vec{k}$ is in Å^{-1}, and $\alpha = 0.2624$ (eV Å^2)$^{-1}$. The exponential in Eq. (5) is identical to that which appears in the free-electron solution for the triangular barrier. On the other hand $P(n, \vec{k})$, which relates the details of the electronic band structure to the emission process, differs drastically from a free-electron formulation. Here $P(n, \vec{k})$ is a function of (a) the mixing coefficients which define the superposition of the $3d$ eigenstates; and (b) the $\vec{G}_\parallel$th Fourier components of the projections on the emission surface of the atomic $3d$ orbitals and their derivatives. The combination of (a) and (b) gives rise to the important result that there is an insignificant amount of tunneling in the [001] direction from states having $|\vec{k}_\parallel| = 0$. In contrast to this, free-electron theory predicts maximum emission from states lying along the normal tunneling direction for all crystallographic planes.

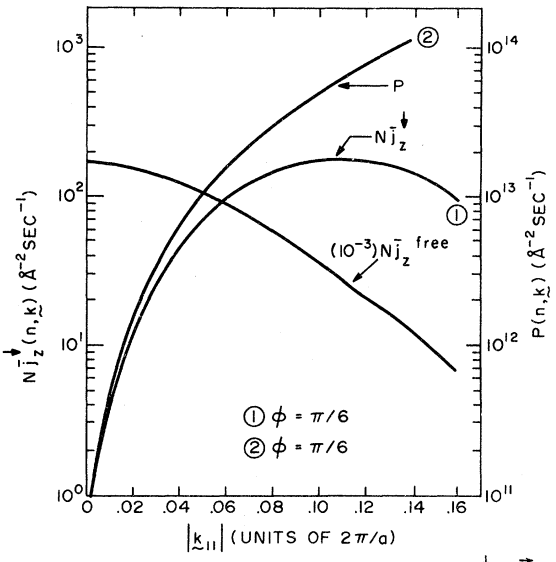

FIG. 1. Averaged emitted current density $\bar{j}_z^{\uparrow}(n,\vec{k})$ and its pre-exponential $P(n,\vec{k})$ versus $|\vec{k}_{||}|$ for polar angle $\varphi \equiv \tan^{-1}(k_y/k_x) = \pi/6$. N denotes the number of primitive unit cells in the unit volume. The $3d$ minority-spin states belong to the large hole pocket (at X) of the Fermi surface. The figure illustrates that the $3d$ current around the [001] direction vanishes from states having $|\vec{k}_{||}| = 0$. On the other hand, the free-electron formulation predicts maximum emission for states with zero transverse momentum.

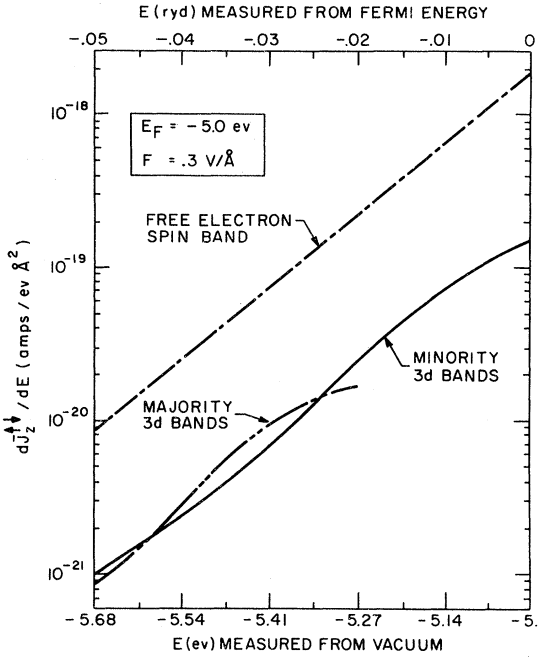

FIG. 2. Energy distribution for field-emitted electrons around the [100] axis. The $3d$ distributions are compared with a free-electron–like $4s$-p band with bottom at Γ_0 (13 eV below vacuum). There are no majority-spin bands above $E \simeq -0.02$ Ry (top scale). The structure in the $3d$ distributions results from the complicated topology of the constant-energy surfaces.

Figure 1 illustrates this situation. Including the spatial symmetries of the $3d$ eigenfunctions in $P(n,\vec{k})$ and $\bar{j}_z^{\uparrow(\downarrow)}(n,\vec{k})$ also leads to anisotropic effects which depend on the orientation of $\vec{k}_{||}$ in the emission plane. Therefore, given a state $(n,\vec{k})$, the nature of the anisotropies will vary according to the crystallographic indices of the emission surface.

The macroscopic current density at the detector, $\bar{J}_z^{\uparrow(\downarrow)}$, is obtained by summing the individual contributions $\bar{j}_z^{\uparrow(\downarrow)}(n,\vec{k})$ over occupied $3d$ states:

$$\bar{J}_z^{\uparrow(\downarrow)} = e\sum_{n,\vec{k}} N^{\uparrow(\downarrow)}(n,\vec{k})\bar{j}_z^{\uparrow(\downarrow)}(n,\vec{k}). \quad (6)$$

$N^{\uparrow(\downarrow)}(n,\vec{k})$, the number of electrons in the spin state $(n,\vec{k})$, is given by the Fermi-Dirac distribution for $T = 0$°K, When the sum over discrete $\vec{k}$ is converted to an integral over the constant-energy surfaces, $\sigma_n^{\uparrow(\downarrow)}(E)$, $\bar{J}_z^{\uparrow(\downarrow)}$ becomes

$$\bar{J}^{\uparrow(\downarrow)} = \frac{eV}{(2\pi)^3} \times \sum_n \int_{-\Phi-E_F}^{-\Phi} dE \int \frac{\bar{j}_z^{\uparrow(\downarrow)}(n,\vec{k})}{|\nabla_{\vec{k}} E_n^{\uparrow(\downarrow)}(\vec{k})|} d\sigma_n^{\uparrow(\downarrow)}(E). \quad (7)$$

Since Φ designates the work function, the energy integral above is taken from the bottom of the conduction band to the Fermi energy.

The $3d$ band structure of Ni was included in the evaluation of Eq. (7) in three distinct ways: (1) The topology of the constant-energy surfaces was determined numerically and used without approximation to define $\sigma_n^{\uparrow(\downarrow)}(E)$ for the integration; (2) at each point $\vec{k}$ on the surface, the density of states, proportional to $|\nabla_k E_n^{\uparrow(\downarrow)}(\vec{k})|^{-1}$, was computed numerically; (3) the corresponding eigenfunctions $|n,\vec{k}\rangle$ were derived, and $\bar{j}_z^{\uparrow(\downarrow)}(n,\vec{k})$ evaluated. The entire integration was performed numerically with a final accuracy of at least two significant figures.

Figure 2 shows the energy distributions, $d\bar{J}_z^{\uparrow(\downarrow)}/dE$, for majority ($\uparrow$) and minority spin ($\downarrow$) electrons. We note that $d\bar{J}_z^{\uparrow(\downarrow)}/dE$ for the $3d$ bands are an order of magnitude less than $d\bar{J}_z^{free}/dE$, the contribution of the $4s$-p bands—although both appear to have approximately the same exponential dependence on energy. One immediately concludes that field emission from a (100) plane of Ni is largely due to the free-electron–like $4s$-p

band. This result suggests why the free-electron Fowler-Nordheim theory adequately describes most experimental observations on field emission from d-band transition metals. Further note that the small amount of structure exhibited by the majority-spin $3d$ distribution at an energy E is quite similar to that in the minority-spin distribution at $E+\Delta E_{\text{exch}}{}^{(d)}=E+0.03$ Ry. Since the shapes of the minority and majority constant-energy surfaces are identical at energies $E+\Delta E_{\text{exch}}{}^{(d)}$ and E, respectively, we deduce that the structure is a manifestation primarily of the surface topology.

Finally, the energy distributions of Figure 2 were integrated yielding $\bar{J}_{3d}{}^{\uparrow(\downarrow)}$ and $\bar{J}_{4s-p}{}^{\uparrow(\downarrow)} \equiv \bar{J}_{\text{free}}{}^{\uparrow(\downarrow)}$. (The z subscript is now omitted for the sake of simplicity.) The resulting spin polarization given by Eq. (1) is -4%, the preferred spin direction being parallel to the magnetization of the crystal. The negative sign is a consequence of the fact that the majority-spin $3d$ bands do not contribute to the Fermi surface. The tunneling process—which discriminates in favor of Fermi-energy electrons—selectively emits the ($\downarrow$) $3d$ electrons from the partially-filled minority spin bands.

Possible sources of error in the calculation include: (i) over-estimating the current from the $4s$-p bands when using the free-electron approximation[13]; (ii) neglecting depolarization (spin-flipping) effects such as electron-magnon scattering[14] and spin-dependent transmission through the surface potential barrier; and (iii) depolarization due to inhomogeneities in the external magnetic field.[3,15] Nevertheless, it is felt that the ESP obtained above represents good agreement with the -10% reported by Gleich, Regenfus, and Sizmann[3] and consequently lends further evidence to the validity of the SWS itinerant band model. It is suggested that the sign discrepancy of ESP found in the photoemission experiments[1,2] originates in the use of polycrystalline samples rather than the inapplicability of the SWS band model. The results of Gleich, Regenfus, and Sizmann—which show the sign and magnitude of the ESP to be a strong function of crystallographic direction—gives evidence for this fact.

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