fits stem from the assumption that the number of Bloch walls pinned at temperature $T$ is described by a Lorentzian function. Although this assumption is quite plausible, it would be reckless to view the agreement as a verification of the theory.

If the spacing of the Bloch walls at $T_c$ is $\approx 10^{-4}$ cm for a sample with a 1-cm$^2$ cross section, then $n(\kappa_0) \approx 10^4$ walls/cm$^3$. Galt $et$ $al.$ measured the frequency dependence of the initial permeability in magnetite ($\text{Fe}_3\text{O}_4$) and in NiFe$_2$O$_4$, and deduced appreciably different values of $\beta$ for the two samples, 0.406 and 0.026 g cm$^{-2}$ sec$^{-1}$, respectively. If $\beta \approx 1.0$ g cm$^{-2}$ sec$^{-1}$ represents an upper limit on the damping constant, then one could assume that $n/\beta \approx 10^4$ for Dy and Tb samples. We can, therefore, infer from the values of $nf/\beta$ given in Table I that the Tb absorption was nearly on-resonance ($f \approx 1$) in zero applied field, whereas the Dy absorption was far off-resonance ($f \ll 1$) in zero applied field. Apparently, application of the magnetic field in Dy causes changes in $m_0$, $\beta$, and $\kappa_0$ such that the domain walls are more nearly brought into resonance with the sound wave. This possible explanation of the seemingly contradictory zero-field observations is, of course, pure speculation. Further theoretical interpretation must await a careful investigation of the spectral dependence of the attenuation.


Nickel Sulfide—an Itinerant-Electron Antiferromagnet

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(Received 4 March 1974)

Antiferromagnetic nickel sulphide has a small temperature-independent magnetic susceptibility. The ordered moment per nickel atom, measured with neutrons, decreases from 1.45$\mu_B$ to 1.00$\mu_B$ at 4.2 K with a decrease of 0.6% in the Ni-Ni distance. These data indicate a band model for the 3d electrons, making NIS an itinerant-electron antiferromagnet with no local moments. The possibility of local moments in an antiferromagnetic metal is briefly discussed.

The existence of itinerant-electron antiferromagnetism has been established with certainty in only a few materials, notably chromium metal.1 An antiferromagnetic metal is predicted as an intermediate state between the antiferromagnetic insulator and the Pauli-paramagnetic metal as
some parameter such as the lattice constant or
the intra-atomic Coulomb interaction is varied.²
Nevertheless, it does not occur in the most ex-
tensively studied metal-insulator transition sys-
tem (V₁₋ₓCrₓ)₂O₃³ probably because the jump in
the lattice parameter at the transition is such
that the itinerant-electron antiferromagnetic
state would appear in the “lost volume.”⁴ In this
Letter, we present evidence to show that nickel
sulphide is an itinerant-electron antiferromagnet
below its first-order magnetic ordering tempera-
ture⁵ Tᵢ, changing to a metal with a very short
spin-spin correlation time above Tᵢ. Our sus-
ceptibility and neutron-diffraction data below the
transition seem incompatible with the presence
of local moments on the nickel sites, assumed by
previous workers,⁶⁷ although above Tᵢ we con-
firm White and Mott’s interpretation of NiS as a
normal d-band metal.⁶

A semimetallic or metallic conductivity might
be considered as evidence that an antiferromag-
net is itinerant-electron type, but this condition is
insufficient because the conduction electrons
are not necessarily the magnetic electrons. The
conductivity, shown in Fig. 1(a), is substantially
independent of temperature below Tᵢ, in agree-
ment with earlier work.⁷ It would be consistent
with NiS being either a degenerate semiconductor
or a semimetal below Tᵢ, although the electronic-
specific-heat coefficient, γ = 0.9 ± 0.4 mJ/mole
K²,⁹ favors the semimetallic interpretation.

Above Tᵢ the conductivity has a normal metallic
temperature dependence. To show that NiS is
an itinerant-electron antiferromagnet, it is suffi-
cient to show that the magnetic electrons are
itinerant. We do this by pointing out that the sus-
cceptibility is inconsistent with the existence of S
≈ 1 local moments, and that the ordered moment
varies as a function of cell parameter to an ex-
tent highly unlikely if the moments are localized.

Our first evidence in favor of a band model for
the nickel moments is provided by the suscepti-
bility, Fig. 1(b): Both χ∥ and χ⊥ are remarkably
independent of temperature in the antiferromag-
netic phase. If χ∥(T) - χ⊥(T) is fitted with the
standard local-moment model,¹⁰ the extrapolated
Néel temperature would be ~1600 K, implying
unprecedented superexchange interactions. How-
ever, both the nickel moment⁸ and the hyperfine
field on ⁵⁷Fe impurities¹¹ decrease by 10% in the
range 0 < T < Tᵢ, giving an extrapolated Néel
temperature of only ~400 K if local moments are
assumed.¹¹ ¹² The inconsistency vanishes with a
band model. The simplest is the Stoner-Liddard
model.¹³ The intersublattice exchange, which we
assume to be dominant, is determined to be kθ' 
≈ 0.18 eV from the temperature dependence of
the nickel sublattice magnetization and its re-
duced value at T = 0, θ₀ ≈ 0.7. The perpendicular
susceptibility is given by

χ⊥ = N₀μ² / ℏθ',

where N₀ is the number of electrons in the mag-
netically polarized band (two per nickel atom).
The calculated value is χ⊥ = 3.96 × 10⁻⁶ emu/g, in
good agreement with observation [Fig. 1(b)]. The
parallel susceptibility, unlike the perpendicular
susceptibility, depends on the form of the density
of states which is not yet known with any certain-
ity in NiS. Nevertheless, it should be rather in-
dependent of temperature, and tend to a nonzero
value as T → 0.¹³ The recent theory of Brandt
and Gross¹⁴ for itinerant-electron antiferromagnets
also predicts that χ∥ varies little between
T = 0 and Tₙ provided there is no Hubbard gap at
T = 0. This is consistent with the semimetallic
behavior below Tᵢ. The spin-wave dispersion
relation for NiS was found to be very steep,¹⁵ and
also implies a very high extrapolated Néel tem-
perature if interpreted on a local-moment model.
As it appears this is not the case¹¹ ¹²; the stiff
spin waves should rather be regarded as evidence
for itinerant-electron antiferromagnetism.¹⁵
Our second evidence in favor of a band model follows from neutron-diffraction powder patterns which provided us lattice parameters and magnetic moments for a series of five samples Ni_{1.5}S of differing stoichiometry, 0 < \delta < 0.045. The homogeneity of each of the first four samples was deduced from the width of the specific-heat anomaly at T_f (\approx 10 K) to be better than \pm 0.001 in \delta. The nonstoichiometry takes the form of vacancies on the nickel lattice,^{18} and has the effect of reducing the lattice parameters, particularly in the c direction. This appears rather clearly in Fig. 2(b), although neutron diffraction is not the most precise technique for lattice-parameter determinations. Moreover there seems to be an abrupt decrease in c near \delta = 0.03 at 4.2 K associated with loss of moments [Fig. 2(a)]. The moments are derived from (101) and (111) reflections at 4.2 K, the only ones with any substantial magnet-
TABLE I. Data on five samples of Ni$_{1-\delta}$S.

<table>
<thead>
<tr>
<th>$\delta$</th>
<th>Moment at 4.2 K ($\mu_B$)</th>
<th>Transition temperature (K)</th>
<th>Ni–Ni (Å)</th>
<th>Ni–S (Å)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.000</td>
<td>1.45 ± 0.20</td>
<td>264 ± 5</td>
<td>2.702 ± 0.005</td>
<td>2.462 ± 0.005</td>
</tr>
<tr>
<td>0.007</td>
<td>1.29 ± 0.10</td>
<td>240 ± 5</td>
<td>2.694 ± 0.005</td>
<td>2.398 ± 0.005</td>
</tr>
<tr>
<td>0.015</td>
<td>1.17 ± 0.10</td>
<td>196 ± 5</td>
<td>2.692 ± 0.005</td>
<td>2.397 ± 0.005</td>
</tr>
<tr>
<td>0.024</td>
<td>1.00 ± 0.10</td>
<td>146 ± 6</td>
<td>2.687 ± 0.005</td>
<td>2.393 ± 0.005</td>
</tr>
<tr>
<td>0.035</td>
<td>0.00 ± 0.00</td>
<td>⋯</td>
<td>2.659 ± 0.005</td>
<td>2.383 ± 0.005</td>
</tr>
</tbody>
</table>

Properties of the antiferromagnetic state from measurements in the paramagnetic state because they are different phases, separated by a first-order transition.

We have argued here that nickel sulphide is an itinerant-electron antiferromagnet because the magnetic electrons are delocalized, and must be described by a band model. However, the breakdown of the Mott insulating state does not inevitably require the disappearance of local, Hund's-rule moments. When the electrons are delocalized for only a small fraction of the time, remaining highly correlated for most of the time, an antiferromagnetic metal with quite different magnetic properties from NiS results. A possible example is FeS, where the low-lying $a$-spin electrons are localized, and perhaps only the single $\beta$-spin electron at the top of the band is itinerant. A transition from local-moment to band-moment behavior in the antiferromagnetic metal with decreasing lattice parameter is conceivable, though no example is known to the authors.

We wish to thank M. Cyrot, G. Czjzek, M. T. H. Hutchings, and R. Natoli for their helpful comments.

9J. M. D. Coey and R. Brusetti, to be published.
12A similar temperature dependence of the hyperfine field measured directly on $^{64}$Ni has been found by J. Fink and G. Czjzek, to be published.