

FIG. 2. Recordings of the n=1 line for change in $\langle N \rangle$ in the vicinity of 38. A current of 2.18 mA through the biasing resistor *R* corresponds to $\Phi_0 \omega_D$.

tion and then, by standard Fourier inversion, the correlation function for the voltage across the contact. Evaluation of this correlation as a function of $\langle N \rangle$ by varying temperature, ambient magnetic field, and contact geometry will then be possible.

Two extreme models of the flux motion are the following: (1) The contact becomes filled with (N-1) vortices, and on introduction of the *N*th vortex, all *N* are released, and (2) all *N* flux quanta cross the contact at once when some barrier for field penetration is exceeded. Very likely this question can be resolved by a careful Fourier analysis of the entire spectrum. For example, if the first model were correct, then the line at n=N, $V_0 = \Phi_0 \omega_D / 2\pi$ should be proportionately larger than the remainder of the nearby spectrum. No definitive statement can be made from the present experimental data.

The lack of symmetry in the spectrum, as shown in Fig. 1(a), is probably due to an asymmetry in the contact geometry and hence to an asymmetry in the barrier to vortex formation. This should be sensitive to reversals in the ambient magnetic field.

⁴This modification of the current-velocity function is indicated in Ref. 1 and experimentally shown in Ref. 2.

 ${}^{5}\text{R.}$ A. Kamper, in Symposium on the Physics of Superconducting Devices, 1967 (unpublished). We have observed that the linewidths at 30 MHz for 3.6- and 25.6- $\mu\Omega$ resistors vary approximately as R, and the temperature dependence of this width is presently being measured.

⁶The critical current can be measured by an rf method similar to that described for superconducting rings in Ref. 2 and is described by A. H. Silver, in Symposium on the Physics of Superconducting Devices, 1967 (unpublished).

LOW-ENERGY INTERBAND TRANSITIONS AND BAND STRUCTURE IN NICKEL

J. Hanus, J. Feinleib, and W. J. Scouler

Lincoln Laboratory,* Massachusetts Institute of Technology, Lexington, Massachusetts (Received 22 May 1967)

Thermal modulation experiments in the low-energy region favor a reordering of the bands at the L point in the Brillouin zone in nickel.

Low-energy interband transitions detected by thermally modulated reflectivity^{1,2} lend new support to a reordering of the bands at the Lpoint in Ni. We show that this modification brings the band structure at the Fermi surface into better agreement with all relevant experiments.

The pulsed-current modulated reflectivity $(\Delta R/R)$ data were obtained from 0.1 to 10 eV on liquid-N₂-cooled Ni films as shown in Fig. 1. We will concentrate here on the interpretation

of the low-energy part from 0 to 2 eV. The structure in this range, which has been attributed to transitions around the L point³ in the Brillouin zone, is resolved by this technique into separate peaks at 0.25 and 0.4 eV and a shoulder at 1.3 eV. The requirements of a model to fit these data must also be compatible with the following experimental results. Saturation magnetization⁴ gives an excess of 0.55 majority spin-up (†) electrons, while a Fermi surface enclosing a net volume correspond-

¹J. E. Zimmerman, J. A. Cowen, and A. H. Silver, Appl. Phys. Letters <u>9</u>, 353 (1966).

²A. H. Silver and J. E. Zimmerman, Phys. Rev. <u>158</u>, 423 (1967).

³A. H. Silver and J. E. Zimmerman, Appl. Phys. Letters 10, 142 (1967).



FIG. 1. The temperature-modulated reflectance $\Delta R/R$, and the reflectance R, for a nickel film from 0.2 to 10 eV.

ing to one electron is deduced from magnetoresistance.⁵ de Haas-van Alphen^{6,7} data indicate a Fermi surface with an open sheet with necks in the (111) direction [neck radius 0.09 \AA^{-1} and $m_1(\text{neck}) = 0.25m_0$ and also a pocket of holes which is interpreted as coming from an $X_{\mathfrak{s}}$ band. The ferromagnetic Kerr effect (FKE)⁸ which in theory distinguishes ↑ and ↓ spin optical transitions, has been $used^{9-11}$ to analyze different models of the electronic structure about the L point. Measurements by Krinchik et al.¹² and Martin et al.¹³ have given qualitatively different results¹⁴ from 0.2 to 1.5 eV [Fig. 2(a)], which lead to different spin assignments to the deduced optical transitions in this range. In view of this, the observed structure from 0.2 to 0.4 eV and the long tail out to 1.4-1.6 eV in the FKE results as well as other poorly resolved optical^{3,15} data are not sufficient to distinguish between different models. All the previous experimental results along with the theoretical estimates of the ferromagnetic splittings¹⁶ of the bands were not sufficient to fix the relative positions of the s-p-like conduction band at L_{2}' , the dband at L_{32} , and the Fermi level.

Previous controversy^{3,9} over the interpretation of the experimental data relevant to the one-particle spectrum of nickel near the Fermi surface was concerned with slight modification of the spacing of these levels, but did not change the ordering calculated for paramagnetic nickel.¹⁷⁻¹⁹ The recent self-consistent interpolation scheme²⁰ of Hodges, Ehrenreich, and Lang (HEL) based on these paramagnetic calculations and incorporating correlation effects achieved a good interpretation of most



FIG. 2. Spectral dependence of $\epsilon_m^{(1)}$. (a) $\epsilon_m^{(1)}$ from experimental ferromagnetic Kerr effect (Refs. 12 and 13). (b) Sketch of the spectral dependence deduced from models A and B. The amplitude of each of the three contributions reflects only the $1/\omega^2$ dependence, but the matrix elements are not calculated.

of the experiments including the magnetic form factor and anisotropy.²¹ However, their band model retains the L-point ordering of the paramagnetic bands $[E(L_2')-E(L_{32})>0]$ and gives a majority spin band (*) similar to that of copper,²² which we shall call model A^{\ddagger} . The possibility of a new ordering $[E(L_2') - E(L_{32}) < 0]$ was considered by Krinchik²³ to account for the FKE. Self-consistent band calculations for the ferromagnetic state^{24,25} have been made recently. Connolly, using a reduced exchange potential,²⁶ obtained a minority spin band with this new ordering. Our optical data favor such an ordering in both up and down spin bands, and we believe this model explains the pertinent data better than previous metals.

Around L and for energies close to the Fermi energy we show in Fig. 3 the effects of the old (A) and new (B) ordering schemes on the majority (\dagger) and minority (\dagger) spin bands, and also take into account the *s*-*d* hybridization



FIG. 3. Band structure of nickel near L for models A and B. Energies are in electron volts.

of the Q_{-} states. The position of the Fermi level (above L_{32}^{\dagger} and below L_{32}^{\dagger}) in both models A and B is set by the net spin moment. The relative position of $E_{\mathbf{F}}$ and L_{2}' is determined by de Haas-van Alphen data^{6,7}: The presence of (111) neck indicates that L_{2}' is below $E_{\mathbf{F}}$ in A^{\dagger} ; the absence of L holes in these data places L_{2}' below $E_{\mathbf{F}}$ in B^{\dagger} . These considerations do not fix the energy spacings in either model so HEL could explain most of the experimental data by an appropriate choice of these gaps within model A, while Krinchik could claim a better fit with his data using model B.

The fit of our new optical data with model $A \downarrow$, as shown in Fig. 3, requires $L_{32} \downarrow$ to be 0.4 eV above E_F as compared with 0.24 eV in HEL and, therefore, would give too many unpaired spins.^{9,27} If we use $B \downarrow$, even with a vanishing gap $[E(L_{32})-E_F \sim 0 \text{ eV}]$, model A^{\dagger} still gives an unreasonably large exchange splitting if the 1.3-eV optical structure occurs within the majority spin band as is commonly assumed. Model $B \downarrow$, unlike model $A \downarrow$, has the great advantage of ruling out any possibility of holes around L. In addition, using the interpolation scheme of Hodges,²⁸ for this model, the possibility of holes at X_2 is ruled out; this may explain why careful de Haas-

van Alphen (dHvA) investigations⁷ have not revealed previously predicted X_2 hole pockets.

Phillips⁹ considered a model (Ref. 9, model c) which would give a dHvA neck at L_{32} as we have in B^{\dagger} . In his model, this neck came from the Q_+ band rather than the Q_- because L_{2}' was above L_{32} and gave Q_{-} a negative curvature. But the transverse mass was far too large since the $L_{32}Q_+W_1'$ band is flat, and therefor Phillips ruled out the possibility of d necks. Our B^{\dagger} model does not have a large-mass neck because s - p character is acquired through hybridization. Furthermore, the longitudinalmass parameter m_{l} ,²⁹ required by Joseph and Thorsen⁶ to fit their de Haas-van Alphen data, is much greater than m_1 for the noble metals and therefore suggests a departure from Cu-like necks. Model B^{\dagger} brings in a new feature: the possibility of a low-energy optical transition $(Q_+ \uparrow \neg Q_- \uparrow)$ which we believe is the one observed at 0.25 eV. In Fig. 2(a) we show $\epsilon_m^{(1)}$, the off-diagonal absorptive part of the dielectric constant derived from the FKE data. Figure 2(b) shows the qualitative features predicted for the FKE effect by models A and B. A more quantitative comparison requires a knowledge of the three matrix elements involved. Both sets of FKE data agree only in the very limited range 0.25-0.3 eV.14 The important features which are required to fit the models are the negative maxima, but these peaks fall on either side of the common range of agreement. While one set of data suggests a positive peak below 0.25 eV as in our model B, the other data suggest a negative peak more like model A. Therefore the available FKE data in themselves do not warrant a distinction between the models.

From our model we see that the d exchange splitting at L is about 0.3 eV and the exchange splitting of the conduction levels is about 0.7 eV. Near the Fermi surface the average character of the d bands is $85\% T_{2g}$ and $15\% E_g$. At L the character is about 75 and 25%, respectively. Since the exchange energy of T_{2g} orbitals¹⁹ is about four times larger than the E_{g} splitting, the average exchange at the Fermi surface should be about 0.33 eV. Herring¹⁶ has estimated the exchange to be 0.25 eV and has argued against a value larger than 0.35 eV. Our estimate falls in this range, as does HEL's estimate. The latter was determined mostly by considerations of the magnetic properties of nickel which are dominated by the

region around X. Our model does not substantially modify their interpretation of magnetic properties²⁸ but allows a more satisfactory interpretation of the low-energy optical data.

The analysis of the line shape in our modulated reflectance data gives us additional information about the gap $E(L_{32}) - E_F$. The transition starting at L is cut off when the Λ_s band crosses the Fermi level. Our linewidth would therefore give $E(L_{32}) - E_F \sim 0.05$ eV. Also, as we have found, the temperature effect on the Fermi level would cause this 0.4-eV transition to wash out at higher temperatures. Since this transition does not involve the Fermi surface, it should be sharper but also more sensitive to strain in the sample than the lower energy transition.

A last comment about the spin-orbit splitting is relevant due to the small value of $E(L_{32}^{\downarrow})$ $-E_{\rm F}$. The spin-orbit splitting (~0.1 eV in atomic nickel) will lift the L_{32} degeneracy. In model *B* this could introduce some additional structure at the 0.4-eV transition, but in no way changes the over-all fit of the model since there are no crossing bands.

We would like to acknowledge the very useful discussions we have had with H. Ehrenreich and L. Hodges, and thank them for making available to us their current work on the subject. We appreciate the very stimulating conversations with G. F. Dresselhaus and other colleagues. tions in the \checkmark and \uparrow spin bands. G. S. Krinchik and E. A. Canshina, Phys. Letters 23, 294 (1966), failed to recognize it and ϵ_{m1}^{1} is always of the same sign in their Letter. It is shown by S. Doniach, in <u>Proceedings</u> of the International Colloquium on Optical Properties and Electronic Structure of Metals and Alloys, Paris <u>1965</u> (North-Holland Publishing Company, Amsterdam, 1966), p. 109, that intraband transitions cannot account for the data in nickel.

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¹⁷J. Hanus, Massachusetts Institute of Technology Solid State and Molecular Theory Group, Quarterly Progress Report No. 44, 1962 (unpublished), p. 29. There is a misprint in the table given in that reference, which is responsible for much speculation. On p. 34, $E(X_5)$ should read 0.6054 instead of 0.6254 Ry and give a flat X_5W_1' band. In this reference the potential was chosen to give paramagnetic bands that interpolate smoothly between Cu and paramagnetic Fe [J. H. Wood, Phys. Rev. <u>117</u>, 714 (1960); <u>126</u>, 517 (1962)].

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²⁸We are very much indebted to Dr. L. Hodges for a copy of interpolation program and communication on model B. See also L. Hodges, to be published.

 ${}^{29}m_l/m_0 = 0.65 \pm 0.10$ Ni, 0.177 for Cu, 0.124 for Ag, and 0.122 for Au.

INFINITE SUSCEPTIBILITY WITHOUT LONG-RANGE ORDER: THE TWO-DIMENSIONAL HARMONIC "SOLID"

B. Jancovici

Laboratoire de Physique Théorique et Hautes Energies, Orsay, France* (Received 15 May 1967)

It is pointed out that the classical two-dimensional harmonic "solid" exhibits an infinite generalized susceptibility at low temperature, although there is no long-range order and no phase transition.

It has been rigorously proven that there is no ferromagnetism in the two-dimensional isotropic Heisenberg model.¹ There are, however, strong indications that the magnetic susceptibility becomes infinite below some critical temperature.² It has therefore been speculated that, below this critical temperature, the curve of the magnetization versus the field might have a vertical tangent at zero field, without having a finite discontinuity.

In the present note, it is proven that a very simple soluble model, the classical two-dimensional harmonic "solid," does exhibit this very behavior. Let \vec{K} be a vector of the reciprocal lattice. The analog of the spontaneous magnetization per particle is N^{-1} times the average value $\langle \rho_{\vec{K}} \rangle_0$ of the Fourier component of the density $\rho_{\mathbf{K}}$ in the limit of no external field. The analog of the susceptibility is the linear response $\chi_{\vec{K}}$ of $(1/N)\langle \rho_{\vec{K}} \rangle$ to the static external potential $V \exp(i\vec{K}\cdot\vec{r})$. It is well known that a harmonic system undergoes no phase transition, in any number of dimensions, and also that $(1/N)\langle \rho_{\mathbf{K}}^{\star}\rangle_0$ is zero in one and two dimensions. We shall prove, however, that in two dimensions, $\chi_{\mathbf{K}}^{\perp}$, finite above a critical temperature T_{c} , becomes infinite at and below that temperature.

We consider a two-dimensional square lattice of N particles of mass m which interact through harmonic forces; periodic boundary conditions are assumed. Let \overline{R}_i be the equilibrium positions of the particles and \overline{u}_i the deviations of these positions from their equilibrium values. The \overline{u}_i are linear combinations of the phonon coordinates³ \overline{v}_k :

$$\vec{\mathbf{u}}_{i} = N^{-1/2} \sum_{\vec{\mathbf{k}}} \exp(i\vec{\mathbf{k}}\cdot\vec{\mathbf{R}}_{i})\vec{\mathbf{v}}_{\vec{\mathbf{k}}}.$$
 (1)

The potential energy of the system is $\sum_{\mathbf{k}} \frac{1}{2}m\omega_{\mathbf{k}}^{-2}$ $\times |\overline{\mathbf{v}}_{\mathbf{k}}^{-}|^2$, where $\omega_{\mathbf{k}}^{-}$ is the angular frequency associated with the wave number \mathbf{k} ; and the sums on \mathbf{k} always run over the first Brillouin zone. A component $v_{\mathbf{k}\lambda}$ of $\overline{\mathbf{v}}_{\mathbf{k}}^{-}$ has a Gaussian distribution with a width given by the energy equipartition at temperature T:

$$\frac{1}{2}m\omega_{\vec{k}}^{2}\langle |v_{\vec{k}}\rangle|^{2}\rangle = \frac{1}{2}k_{B}T.$$
(2)

The total internal energy (including the kinetic energy) is $2Nk_BT$. Since this is a regular function of T, there is no phase transition in the the thermodynamic sense.

The Fourier component of the density $\rho_{\vec{K}}$ is