

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.

5R. A. Kamper, in Symposium on the Physics of Superconducting Devices, 1967 (unpublished).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.

 $6$ The 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rings in Ref. 2 and is described by A. H. Silver, in Symposium on the Physics of Superconducting Devices, 1967 (unpublished}.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

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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<sup>1,2</sup> lend new support to a reordering of the bands at the  $L$ point 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- $N_2$ -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  $L$  point<sup>3</sup> 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 magnetization<sup>4</sup> gives an excess of 0.55 majority spin-up  $(*)$  electrons, while a Fermi surface enclosing a net volume correspond-

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

 ${}^{2}$ A. H. Silver and J. E. Zimmerman, Phys. Rev. 158, 423 (1967).

<sup>3</sup>A. 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.<sup>5</sup> de Haas-van Alphen<sup>6,7</sup> data indicate a Fermi surface with an open sheet with  ${\rm necks}$  in the (111) direction [neck radius 0.09  $\mathring{A}^{-1}$  and  $m_{\perp}(\text{neck}) = 0.25 m_{o}$ ] and also a pocket of holes which is interpreted as coming from an  $X_{\sigma}$  band. The ferromagnetic Kerr effect (FKE)<sup>8</sup> which in theory distinguishes  $\dagger$  and  $\dagger$ <br>spin optical transitions, has been used<sup>9-11</sup> spin optical transitions, has been used<sup>9-11</sup> to analyze different models of the electronic structure about the  $L$  point. Measurements by Krinchik et al.<sup>12</sup> and Martin et al.<sup>13</sup> have given qualitatively different results'4 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<sup>3,15</sup> data are not sufficient to distinguish between different models. All the previous experimental results along with the theoretical estimates of the ferromagnetic splittings<sup>16</sup> of the bands were not sufficient to fix the relative positions of the s-p-like conduction band at  $L_2'$ , the d band at  $L_{32}$ , and the Fermi level.

Previous controversy<sup>3,9</sup> 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 scheme<sup>20</sup> 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  $\epsilon_m$ <sup>(1)</sup>. (a)  $\epsilon_m$ <sup>(1)</sup> 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 of the experiments including the magnetic for<br>factor and anisotropy.<sup>21</sup> 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  $($ *i*) similar to that of copper,<sup>22</sup> which we shall call model  $A$ **<sup>***i***</sup>. The pos** $per, ^{22}$  which we shall call model  $A<sup>4</sup>$ . The possibility of a new ordering  $[E(L_2') - E(L_{32}) < 0]$ was considered by Krinchik<sup>23</sup> to account for the FKE. Self -consistent band calculations for the ferromagnetic state<sup>24,25</sup> have been made recently. Connolly, using a reduced exchange potential, '6 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  $L_{32}^{\bullet}$  and below  $L_{32}^{\bullet}$ ) in both models  $A$  and  $B$  is set by the net spin moment. The relative position of  $E_{\mathbf{F}}$  and  $L_{\mathbf{z}}'$  is determined by de Haas-van Alphen data $6,7$ : The presence of (111) neck indicates that  $L_2'$  is below  $E_F$  in  $A^*$ ; the absence of L holes in these data places  $L_{\mathbf{z}}'$  below  ${E}_{\mathbf{F}}$  in  $B$   $\!{\bullet}$ . 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  $A^*$ , as shown in Fig. 3, requires  $L_{32}^*$  to be 0.4 eV above  $E_F$  as compared with 0.24 eV in HEL and, therefore, would give too many unpaired spins.<sup>9,27</sup> If we use  $B^*$ , even with a vanishing gap  $[E(L_{32})-E_{\overline{F}}\sim 0 \text{ eV}]$ , model  $A<sup>†</sup>$  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  $B^*$ , unlike model  $A^*$ , has the great advantage of ruling out any possibility of holes around  $L$ . In addition, using the interpolation scheme of Hodges,<sup>28</sup> for this model, the possibility of holes at  $X<sub>2</sub>$  is ruled out; this may explain why careful de Haasvan Alphen (dHvA) investigations<sup>7</sup> have not revealed previously predicted  $X<sub>2</sub>$  hole pockets.

Phillips<sup>9</sup> considered a model (Ref. 9, model c) which would give a dHvA neck at  $L_{32}$  as we have in  $B<sup>*</sup>$ . 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 there for Phillips ruled out the possibility of  $d$  necks. Our  $B^*$  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 Thorsen<sup>6</sup> to fit their de Haas-van Alphen data, is much greater than  $m<sub>l</sub>$  for the noble metals and therefore suggests a departure from Cu-like necks. Model  $B^*$  brings in a new feature: the possibility of a low-energy optical transition  $(Q_+^{\dagger} + Q_-^{\dagger})$  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. <sup>A</sup> 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.<sup>14</sup> 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.<sup>7</sup> 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  $75^{\circ}$  and  $25\%$ , respectively. Since the exchange energy of  $T_{2g}$  orbitals<sup>19</sup> is about four times larger than the  $E_{\text{g}}$ splitting, the average exchange at the Fermi surface should be about 0.33 eV. Herring<sup>16</sup> 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 properties<sup>28</sup> 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  $E(L_{32}^{\dagger})-E_{\mathbf{F}}$ . The transition starting at L is cut off when the  $\Lambda_s^*$  band crosses the Fermi level. Qur linewidth would therefore give  $E(L_{32}^{\dagger})-E_{\overline{F}}\sim 0.05$  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  $E(L_{32}^{\dagger})$  $-E_F$ . 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.

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 $^{9}$ J. C. Phillips, Phys. Rev. 133, A1020 (1964); J. C. Phillips and L, F. Mattheiss, Phys. Rev. Letters 11, 556 (1963).

 $^{10}$ B. R. Cooper and H. Ehrenreich, Solid State Commun. 2, 171 (1964); B. R. Cooper, Phys. Rev. 139, A1506 (1965); B. R. Cooper, H. Ehrenreich, and L. Hodges, in Proceedings of the International Conference on Magnetism, Nottingham, England, 1964 (The Institute of Physics and the Physical Society, London, 1965), p. 110.

 $<sup>11</sup>H$ . Ehrenreich, in Proceedings of the International</sup> Colloquium on Optical Properties and Electronic Structure of Metals and Alloys, Paris, 1965 (North-Holland Publishing Company, Amsterdam, 1966), p. 109.

<sup>12</sup>G. S. Krinchik and R. D. Nuralieva, Zh. Eksperim. i Teor. Fiz. 36, 1022 (1956) [translation: Soviet Phys.-JETP 9, <sup>729</sup> (1959)]; G. S. Krinchik, J. Appl. Phys. 35, 1089 (1964); G. S. Krinchik and G. M. Nurmukhamedov, Zh. Eksperim. i Teor. Fiz. 48, 34 (1965) [translation: Soviet Phys. - JETP 21, 22 (1965)].

<sup>13</sup>D. H. Martin, S. Doniach, and K. J. Neal, Phys. Letters 9, <sup>224</sup> (1964); D. H. Martin, K.J. Neal, and T.J. Dean, Proc. Phys. Soc. (London) 86, 605 (1965).

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 $^{16}$ C. Herring, in Magnetism, edited by G. T. Rado and H. Suhl (Academic Press, Inc., New York, 1966), Vol. 4, p. 144.

 $^{17}$ J. 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,  $E(X<sub>5</sub>)$  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. 117, 714 (1960); 126, 517 (1962)].

<sup>\*</sup>Operated with support from the U. S. Air Force.

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<sup>&</sup>lt;sup>7</sup>D. C. Tsui and R. W. Stark, Phys. Rev. Letters  $17$ , 871 (1966); Bull. Am. Phys. Soc. 12, 287 (1967).

<sup>&</sup>lt;sup>18</sup>L. F. Mattheiss, Phys. Rev. 134, A970 (1964).  $^{19}$ E. C. Snow, J. T. Waber, and A. C. Switendick, J. Appl. Phys. 37, 1342 (1966). It is shown here that the relative position of  $L_{32}$  and  $L_2'$  is related to the electronic configuration of nickel.

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 $^{25}$ J. W. D. Connolly, thesis, University of Florida, 1966 (unpublished); Bull. Am. Phys. Soc. 12, 135 (1967); and private communication.

 $^{26}$ W. Kohn and L. J. Sham, Phys. Rev. 140, A1133 (1965).

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 $28$ We 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. m that this is not required.<br>very much indebted to Dr. L. Hodges for a<br>terpolation program and communication on<br>See also L. Hodges, to be published.<br> $= 0.65 \pm 0.10$  Ni, 0.177 for Cu, 0.124 for Ag<br>for Au.

 $m_l/m_0$ and 0.122 for Au.

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

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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.<sup>1</sup> There are, however, strong indications that the magnetic susceptibility becomes infinite below some critical temperature.<sup>2</sup> 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  $\tilde{K}$  be a vector of the reciprocal lattice. The analog of the spontaneous magne tization per particle is  $N^{-1}$  times the average value  $\langle \rho_K^* \rangle_0$  of the Fourier component of the density  $\rho_K^+$  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  $V^{\bullet}_{exp}(i\vec{k}\cdot\vec{r})$ . It is well known that a harmonic system undergoes no phase transition, in any number of dimensions, and also that  $(1/N)\langle \rho_K^2 \rangle_0$  is zero in one and two dimensions. We shall prove, however, that in two dimensions,  $\chi_{\mathbf{K}}$ , 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  $\vec{R}_i$  be the equilibrium positions of the particles and  $\tilde{u}_i$ , the deviations of these positions from their equilibrium values. The  $\bar{u}_i$  are linear combinations of the phonon coordinates<sup>3</sup>  $\vec{v}_{k}$ :

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

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

$$
\frac{1}{2}m\,\omega_{\mathbf{k}}^2\langle|\,\nu_{\mathbf{k}}^2\,\rangle|^2\rangle=\frac{1}{2}k_{\mathbf{B}}T.\tag{2}
$$

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