perature-dependent width.

The first two models which predict, respectively, ~50% and ~0% reduction in the exchange splitting slightly above T_C must be resolved theoretically. The third model can be checked experimentally by careful measurements of the temperature dependence of the width of an isolated surface state on Ni close to the Fermi energy.

The measured splitting of the exchange-split surface states at low temperatures is within experimental uncertainty equal to the measured bulk splitting. We can estimate the possible deviation of the local magnetic moment at the surface compared to the bulk using our data and a simple model. The surface state is assumed to decay exponentially into the bulk and only the outer surface layer is allowed to have a different magnetic moment. If the surface state is totally localized to the first layer then the uncertainties in the surface and bulk measurements produce a ±19% possible deviation in the local magnetic moment at the surface compared to the bulk. If the exponential decay length of the surface state is (a) one, (b) two, or (c) three layers then the possible deviation grows to (a) ~22%, (b) ~30%, and (c) ~35%. Most calculations for Ni give a decay length of states near the Fermi energy of from one to three layers.[14,15]

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First-Principles Calculation of Diamagnetic Band Structure

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For a simple nontrivial model potential, the full quantum mechanics of the Bloch electrons in rational magnetic fields is reduced to a one-dimensional eigenvalue problem and the exact diamagnetic band structure is calculated from first principles. Agreements with and deviations from the predictions of the semiclassical Onsager dynamics are found and discussed.

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In spite of considerable efforts[1,2] a first-principles justification for the so-called semiclassical method for Bloch electrons in a magnetic field, i.e., for the use of the effective Hamiltonian

$$H_{\text{eff}}=E_n(\vec{p}/\hbar-e\vec{A}/\hbar c) \tag{1}$$

for a single band with dispersion $E_n(\vec{k})$, is still lacking. On the other hand, this semiclassical Peierls-Onsager scheme has, over the last three decades, been used with the greatest success to extract precise band data from the different magneto-oscillatory effects for practically all materials. For a review see Shoenberg.[3] On the basis of previous work by our group,[4-6] we have succeeded for the first time in calculating from first principles the complete spectrum of Bloch electrons in a magnetic field; we find magnetic subbands which are generally in excellent agreement with the semiclassical energy levels, though only for nondegenerate bands. Thus our precise numerical results will illustrate both the usefulness and the limits of the semiclassical method.

We start from the free-electron Hamiltonian in the symmetric gauge $\vec{A}=\frac{1}{2}B(-y,x,0)$:

$$H_0=\frac{1}{2m}\left[\left(p_x+\frac{eB}{2c}y\right)^2+\left(p_y-\frac{eB}{2c}x\right)^2+p_z^2\right]=\tfrac{1}{2}\hbar\omega_c[(\alpha^{-1/2}p_x/\hbar+\tfrac{1}{2}\alpha^{1/2}y)^2+(\alpha^{-1/2}p_y/\hbar-\tfrac{1}{2}\alpha^{1/2}x)^2]+p_z^2/2m \quad (2)$$

with $\omega_c=eB/mc$ and $\alpha=eB/\hbar c$ as usual.

Now apply the canonical transformation

$$P=\alpha^{-1/2}p_x/\hbar+\tfrac{1}{2}\alpha^{1/2}y,\quad p=\alpha^{-1/2}p_x/\hbar-\tfrac{1}{2}\alpha^{1/2}y,\quad Q=-\alpha^{-1/2}p_y/\hbar+\tfrac{1}{2}\alpha^{1/2}x,\quad q=\alpha^{-1/2}p_y/\hbar+\tfrac{1}{2}\alpha^{1/2}x;$$
$$\alpha^{1/2}y=P-p,\quad \alpha^{1/2}x=Q+q, \quad (3)$$

which preserves the canonical commutation rules $[Q,P]=i$ and $[q,p]=i$; all other commutators vanish. Then

$$H_0=\tfrac{1}{2}\hbar\omega_c[P^2+Q^2]+p_z^2/2m. \quad (4)$$

Introducing as the simplest nontrivial example a two-dimensional square lattice potential in the x-y plane with only two Fourier components,

$$V(x,y)=2V_0(\cos Gx+\cos Gy),\quad G=2\pi/a,$$

we obtain with (4), omitting the trivial motion in the z direction altogether, the equivalent Hamiltonian

$$H_{eq}(P,p;Q,q)=H_0+V=\tfrac{1}{2}\hbar\omega_c[P^2+Q^2]+2V_0\{\cos[\eta^{1/2}(P-p)]+\cos[\eta^{1/2}(Q+q)]\}, \quad (5)$$

where $\eta=G^2/\alpha$. Equation (5) still represents a two-dimensional problem. Presently a reduction to one degree of freedom appears possible only for rational magnetic fields in the well-known sense (cf. Brown[7]), i.e., $e\vec{B}/\hbar c=(2\pi/\Omega)(L/N)\vec{R}$, where $\vec{R}$ is a lattice vector, Ω is the unit-cell volume, and N,L are integers; hence $\eta=2\pi N/L$.

Since $\exp(\pm i\eta^{1/2}p)$ shifts the argument q by $\pm\eta$, L applications of this translation operator shift the q argument of the second cosine in (5) by $2\pi N\equiv 0 \bmod(2\pi)$. This repetitive structure allows a separation *Ansatz*

$$\Phi(Q,q)=\sum_{i=1}^{L}\Psi_i(Q)\psi_i(q)$$

[for the choice of $\psi_i(q)$ see Obermair and Schellnhuber[9]], which reduces (5) to a set of $L\times L$ matrix operators in the (P,Q) degree of freedom.

In the following we treat exclusively the "pure case" $L=1$ in the sense of Ref. 6. The separation then yields a set of one-dimensional equivalent Hamiltonians

$$H(P,Q;\eta;\kappa,\lambda)=\tfrac{1}{2}\hbar\omega_c[P^2+Q^2]+2V_0\{\cos[\eta^{1/2}(Q+\lambda)]+\cos[\eta^{1/2}(P-\kappa)]\}, \quad (6)$$

where the separation constants $\kappa,\lambda\in[-\pi/\sqrt{\eta},\pi/\sqrt{\eta}]$ may be interpreted as the components of a wave vector spanning the magnetic Brillouin zone.[7] For fixed (κ,λ) the eigenvalue problem of (6) can be shown to have a discrete spectrum; variation of the wave vector then produces the entire magnetic subbands.

On the basis of analytic expressions for the matrix elements of (6) with respect to the eigenfunctions of $\frac{1}{2}(P^2+Q^2)$ we have thus calculated the first-principles band structure in a homogeneous field by the variation method to a very high numerical precision.

On the other hand the exact dispersion of the zero-field Hamiltonian

$$H_{per}=-\hbar^2[\partial_x^2+\partial_y^2]/2m+2V_0[\cos Gx+\cos Gy]$$

has been evaluated (two-dimensional Mathieu bands). This allows us to determine analytically

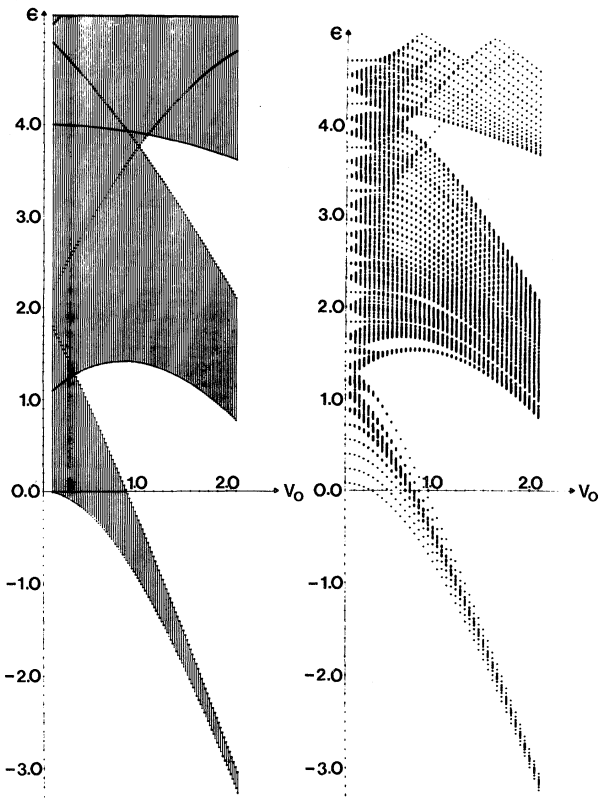

FIG. 1. Left: Zero-field two-dimensional Mathieu band energies ϵ as functions of strength V_0 of the periodic potential. Right: Diamagnetic band structure from first principles for $N=8$ over the same V_0 domain. ϵ and V_0 give the electronic energy and the potential strength in units of 2 eV.

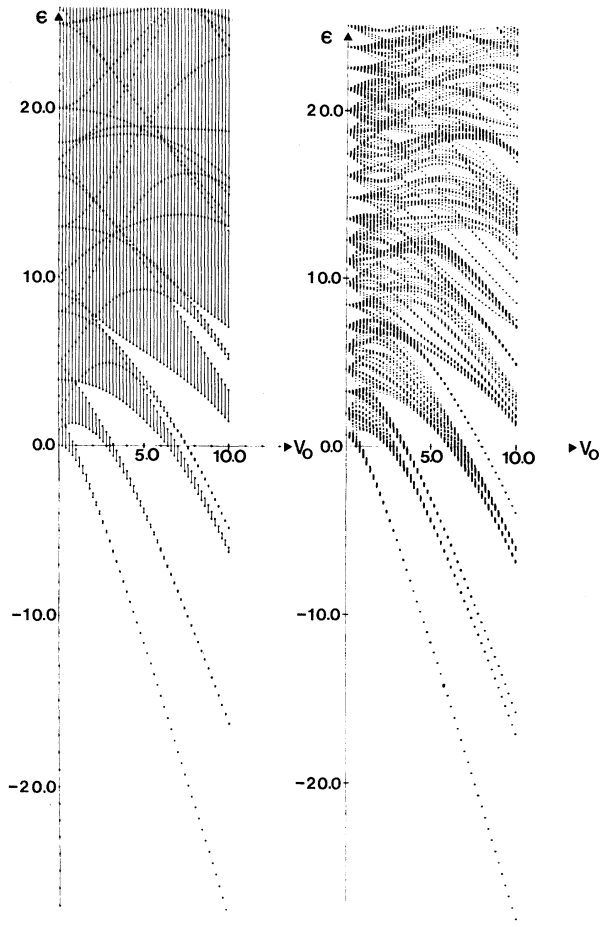

FIG. 2. Same as Fig. 1, but out to higher strengths of the periodic potential and for $N=1$. Notice the splitting of doubly degenerate bands by the magnetic field.

both the "naive" semiclassical spectrum, resulting from the Bohr-Sommerfeld quantization of the magnetic k-space orbits, and, with the aid of previously developed special methods,[6] the quantum-mechanical Peierls-Onsager spectrum, i.e., the eigenvalues of H_{eff} of Eq. (1).

Figures 1 and 2 illustrate the comparison of the zero-field bands with the exact spectrum and of the latter with the semiclassical spectrum. They contrast stability maps of the Mathieu bands with those of the first-principles magnetic subbands as a function of the periodic potential strength V_0 for two values of the magnetic field: $N=8$ ($B\sim 10^7$ G) and $N=1$ ($B\sim 10^8$ G). The pure discrete Landau levels at $V_0=0$ are first strongly broadened by the potential (Landau regime, cf. also Neumann and Rauh[9]), overlap for larger V_0 (regime of magnetic breakdown), and finally separate again into narrow magnetic subbands, which are grouped together to Mathieu bandlike complexes (Onsager regime).

Position and fine structure of these complexes reflect properties of the corresponding Bloch bands: Nondegenerate bands, e.g., the lowest one in Figs. 1 and 2, are split into precisely N subbands in the case of our special rational fields, which—as a rule—lie within the edges of the zero-field band. This agrees fully with group theory and the effective-Hamiltonian concept.

Figure 3 contrasts the first-principles calculation of the magnetic fine structure of the lowest (nondegenerate) Mathieu band with the results of the quantum-mechanical treatment of the Peierls-Onsager effective Hamiltonian H_{eff} for a representative choice of parameters. Shifted somewhat in absolute position, the two spectra nevertheless show excellent agreement with respect to their intrinsic structure (distances and widths).

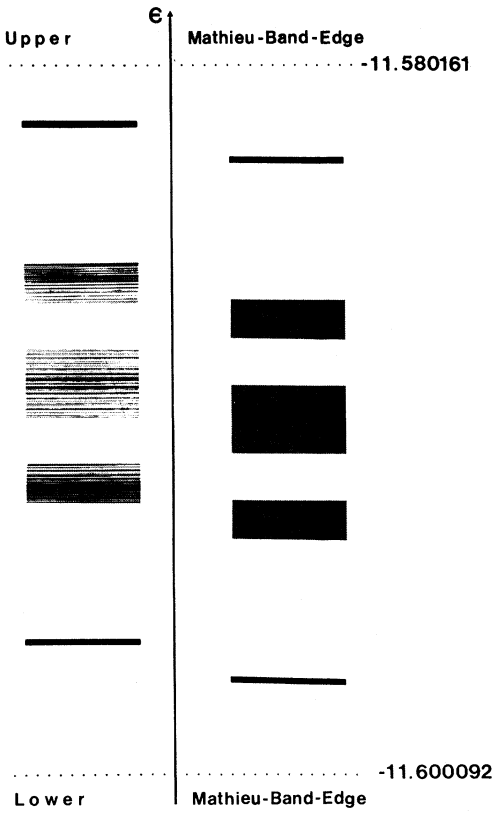

FIG. 3. Magnetic fine structure of the lowest Mathieu band for $N=5$ and $V_0=5.0$. Left: Exact first-principles fine structure. Right: Spectrum of the Peierls-Onsager one-band effective Hamiltonian in quantum mechanical treatment.

A significant deviation from semiclassical behavior, however, occurs in the Onsager regime for degenerate zero-field bands: For sufficiently large periodic potential doubly degenerate Mathieu bands are split by very high magnetic fields into two complexes of N magnetic subbands. For $N=1$ this is clearly visible for the second lowest band in Fig. 2, whereas in Fig. 1 only the onset of the splitting appears for the highest V_0 covered. The two complexes extend well beyond the edges of the zero-field band and can in no way be described by semiclassical methods.

Thus our exact first-principles results serve to confirm the validity of the quantum-mechanical version of the Peierls-Onsager method and —as far as the position of the centers of the magnetic subbands is concerned—also of the semiclassical version, but only for nondegenerate bands.

For degenerate bands the effective-Hamiltonian methods do not work, at least in the very high magnetic fields considered here. Our methods, which can be extended in principle to more complicated crystal potentials and magnetic-field orientations, will permit the evaluation of the diamagnetic band structure in all cases. A detailed account will be given in a forthcoming paper.

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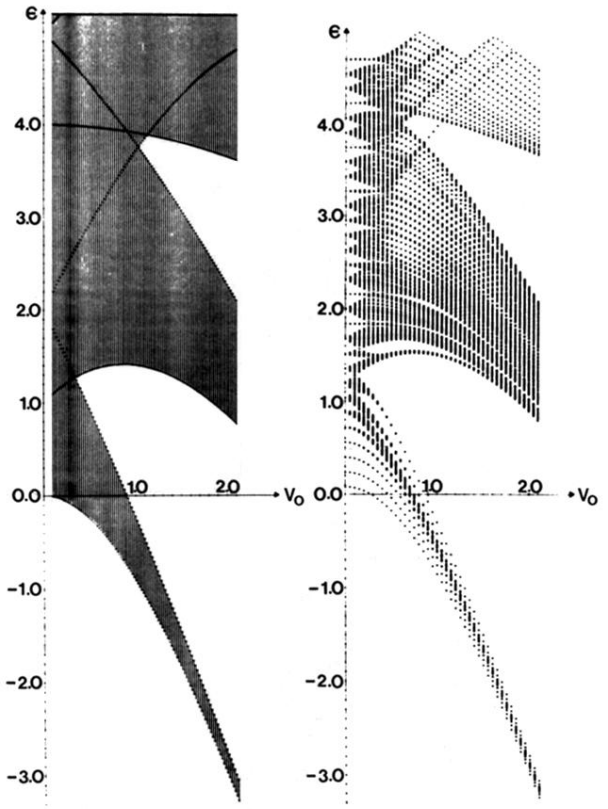

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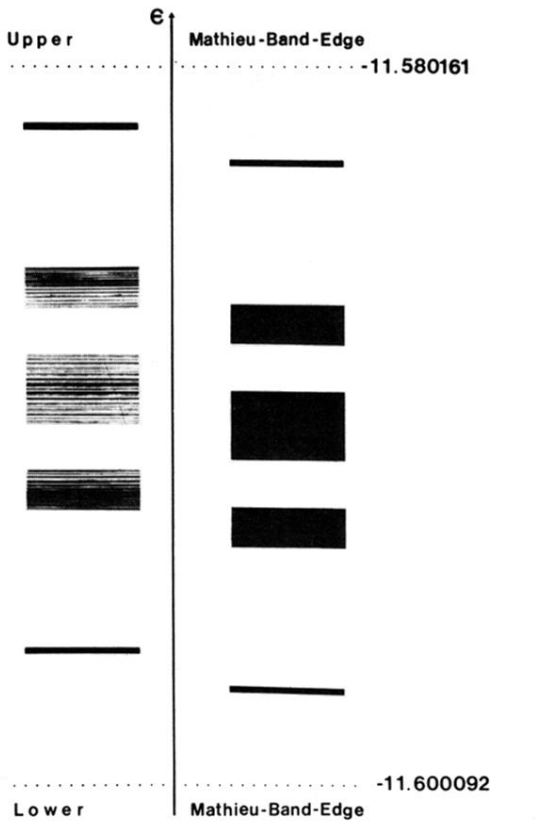

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