Density Waves in High Magnetic Fields: A Metal-Insulator Transition

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(Received 26 August 1985)

The effect of high magnetic fields on the electronic structure of very anisotropic systems with charge- or spin-density waves is examined. In materials where there is lack of perfect nesting at the Fermi surface the density wave produces small electron and hole pockets. Calculations show that the presence of a high magnetic field in certain directions destroys these pockets, opens a gap in the electronic spectrum at the Fermi level, and leads to a metal-insulator transition. This effect has been recently observed in the low-temperature charge-density-wave phase of $NbSe_3$.

PACS numbers: 75.20.Ck, 71.45.Gm

Niobium triselenide is a very anisotropic solid, regarded as an example of a quasi-one-dimensional (Q1D) system. Its monoclinic crystal structure, with 24 atoms in the primitive unit cell, consists of a set of weakly coupled linear chains that run parallel to the b axis.[1,2] The large anisotropy of the system is reflected in its transport properties: The ratio of mobilities along the b and c axes of the crystal is larger than 10. There are two phase transitions[3] in $NbSe_3$ associated with charge-density waves (CDW), at temperatures $T_1 = 144$ K and $T_2 = 59$ K.

It has been shown recently that a high magnetic field, applied *perpendicular* to the niobium chains, i.e., perpendicular to the b axis, couples strongly to the static CDW structure of the low-temperature phase.[4] A large magnetoresistance (which rapidly decreases as the temperature approaches T_2) and an unusual Hall effect show that (1) the observed behavior is not caused by normal galvanomagnetic effects, and (2) the high magnetic field causes a substantial decrease in the number of carriers, i.e., a considerable obliteration of the Fermi surface. Recently,[5] the dependence of the transport properties on magnetic field orientation has been measured, and the results show that only the component of the magnetic field perpendicular to the b axis is responsible for these Fermi-surface effects, which disappear when the field is parallel to the b axis.

In this contribution we show that a high magnetic field can modify considerably the electronic structure of a Q1D metal with a density wave (DW), either a CDW or a spin-density wave (SDW). We conclude that, because of the anisotropy of the system, small electron and hole pockets can be destroyed by the effects of the magnetic field, resulting in a better effective nesting of the Fermi surface and in a semimetal-semiconductor transition controlled by the strength and the direction of the magnetic field.

Our ideas are related to the recent work of Gor'kov and Lebed,[6,7] who studied the effects of magnetic fields in Q1D systems with a nearly stable SDW ground state: They have shown that in very anisotropic metals the tendency to SDW formation is enhanced by the application of an external magnetic field. Héritier, Montambaux, and Lederer[8] generalized the theory and included the magnetic field dependence of the Q vector of the SDW, and suggested that in some systems a sequence of first-order transitions could be observed as the magnetic field increases. Friedel[7] discussed these models in semiclassical terms, valid only for weak magnetic fields, and argued that the analysis should be valid for nearly stable CDW systems as well.

We consider the case of stable DW systems, even in the absence of an applied magnetic field. We assume a tetragonal lattice with constants $a_1 \ll a_2 = a_3$. The one-electron energies in the tight-binding approximation in zero magnetic field are given by

$$\epsilon(\mathbf{k}) = -2t\cos(k_x a_1) - 2t'[\cos(k_y a_2) + \cos(k_z a_3)] - 4t''\cos(k_y a_2)\cos(k_z a_3). \quad (1)$$

Here t and t' are the electron-transfer matrix elements between nearest neighbors in the x direction and in the (y,z) plane, respectively, and t'' connects second-nearest neighbors in the (y,z) plane. The Q1D character of the system results in $t \gg t', t''$. We consider the case of one electron per site. The Fermi surface has then two open sheets (corrugated planes) centered at

$$k_x \approx \pm\pi/(2a_1).$$

For $t''=0$, the Fermi surface has perfect nesting and the system is unstable against the formation of a DW with wave vector $Q = (\pi/a_1, \pi/a_2, \pi/a_3)$. The stable DW is either a CDW or a SDW, depending on whether the electron-phonon or the electron-electron interaction dominates.[9] We characterize the DW by a gap order parameter τ. Because of the extra periodicity induced in the system by the DW, the one-electron state $|\mathbf{k}\rangle$ is mixed with the state $|\mathbf{k}+\mathbf{Q}\rangle$: The matrix element of this mixing is τ, which we assume to be independent of $\mathbf{k}$. The new quasiparticle energy spec-

trum has a gap 2τ at the center of the band (the Fermi level), and the system is therefore a semiconductor.

For $t''\neq 0$ there is no longer perfect nesting of the Fermi surface. If, however, t'' is small enough, a DW with the same wave vector $\mathbf{Q}$ is stable. We consider the case in which

$$t >> t' >> t''. \tag{2}$$

As in the perfect-nesting case, the DW induces a mixing of the $|\mathbf{k}\rangle$ and $|\mathbf{k}+\mathbf{Q}\rangle$ states, but the gap in the spectrum is now given by $2\Delta_g$, where

$$\Delta_g = \begin{cases} \tau - 4t'', & \text{if } \tau/t'' > 4, \\ 0, & \text{otherwise.} \end{cases} \tag{3}$$

In this case, and depending on the value of τ/t'', we have either a semiconductor or a semimetal.

When a magnetic field is applied, a new complication arises. The magnetic field $\mathbf{H}$ quantizes the electron motion in the direction perpendicular to it, and tends to form discrete levels or narrow bands. Problems involving Bloch electrons in a magnetic field have been studied in detail.[10-14] For small values of the field, Onsager's quantization scheme applies, and can be extended to the energy spectrum of a DW system; this approach, however, completely neglects band broadening[11] as well as any tunneling between bands. This last effect is of great importance here since the energy-gap parameter τ may be of the same order of magnitude or smaller than the magnetic energies $\hbar\omega_c$. Under these conditions Onsager's scheme breaks down. It is thus necessary to invoke other methods, which permit the handling of the magnetic field and the DW on the same footing: Harper's generalized equations[13] are ideally suited.

If spin effects are neglected, the Hamiltonian in the presence of a magnetic field in the z direction is given by

$$H = \sum_{nk_\perp\sigma} \epsilon(n,k_\perp) c^\dagger_{nk_\perp\sigma} c_{nk_\perp\sigma} + \tau \sum_{nk_\perp\sigma} \cos(nQ_x a_1) c^\dagger_{nk_\perp\sigma} c_{n(k_\perp+Q_\perp)\sigma} - t \sum_{nk_\perp\sigma} (c^\dagger_{nk_\perp\sigma} c_{(n+1)k_\perp\sigma} + c^\dagger_{nk_\perp\sigma} c_{(n-1)k_\perp\sigma}), \tag{4}$$

where $k_\perp$ ($Q_\perp$) is a two-dimensional vector with components k_y and k_z (Q_y, Q_z). The index n refers to the atomic planes perpendicular to the x direction, σ to the spin index, and the operator $c^\dagger_{nk_\perp\sigma}$ creates an electron in the state $|nk_\perp\sigma\rangle$ of energy $\epsilon(nk_\perp)$ given by

$$\epsilon(nk_\perp) = -2[t' + 2t''\cos(k_z a_3)]\cos(k_y a_2 + \phi n) - 2t'\cos(k_z a_3), \tag{5}$$

where

$$\phi = (|e|Ha_1a_2)/(\hbar c). \tag{6}$$

We have obtained the Hamiltonian (4) in the Landau gauge.

It is clear from (4) and (5) that the magnetic field induces a new periodicity in the system.[14] From the point of view of the mathematics of the problem it is necessary to distinguish between cases in which the periodicities of the lattice and the magnetic field are commensurable or incommesurable. However, as discussed in Refs. 11, 12, and 14, the physical properties of the system are not sensitive to this difference. Without loss of generality we thus consider only the case of "rational" fields.

The energy spectrum of the Hamiltonian (4) consists of a number of magnetic subbands, separated by gaps. The quantity ϕ, defined in (6), is proportional to the number of flux quanta per two-dimensional lattice unit cell (in the plane perpendicular to the direction of the magnetic field), and determines the fine structure of the magnetic subbands. We are not interested in this fine structure, and consider only values of ϕ equal to $(2\pi/N)$, where N is a large integer.[15] The proper scale to measure the magnetic field strength is not ϕ, the magnetic flux, but the magnetic cyclotron energy

$$\hbar\omega_c = (\hbar|e|H)/m^*c,$$

with a cyclotron effective mass m^* corresponding to the bottom of the DW-state conduction band,

$$m^* = (\hbar^2/2a_1a_2)[\tau/t''(t^2-4t'^2)]^{1/2}.$$

We calculated the energy spectrum of the system by means of the transfer matrix method.[12] Because of the mixing of $k_\perp$ and $k_\perp + Q_\perp$, our transfer matrix is of dimension four. For high magnetic fields we obtain a large gap at the Fermi level, in the center of the band. This central gap $2\Delta_g$ is approximately equal to 2τ, even for values of $\tau/t'' < 4$.

We have confirmed these results by a separate and different calculation: We have determined the density of states in the region of interest by a continuous-fraction method.[11,16] The density of states at the center of the band resembles, except for details of the fine structure, the density of states of a system with perfect Fermi surface nesting.

The size of the central gap $2\Delta_g$ decreases monotonically as the magnetic field decreases. Numerical results for a particular case are shown in Fig. 1, and they can be summarized as follows: (A) If condition (2) is satisfied, the size of the central gap Δ_g, measured in units of τ, is sensitive only to the ratios $\alpha = \hbar\omega_c/t''$ and τ/t''. (B) For $\tau/t'' < 4$, the ratio Δ_g/τ is zero for α smaller than a critical value α_c (which depends on τ/t''), and grows continuously from zero

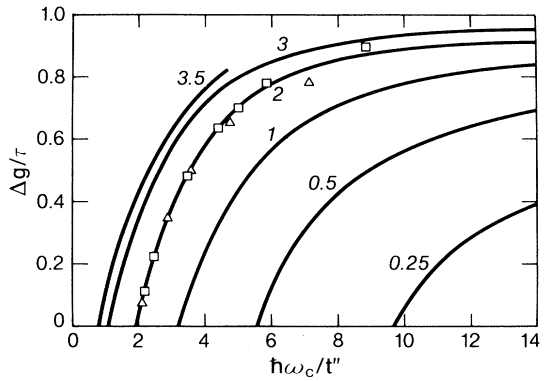

FIG. 1. The value of the energy gap Δ_g in units of the DW parameter τ as a function of the magnetic field strength H, expressed as the ratio of the cyclotron energy $\hbar\omega_c$ to the t'' (departure from nesting) parameter. The lines correspond to various ratios of τ to t'' (values indicated in the graph) and for $t=1$ and $t'=0.2$. For the case of $\tau/t''=2$ we have calculated values of $t'=0.1$ (squares) and $t'=0.3$ (triangles). All curves exhibit a semimetallic region of zero gap, and saturation of Δ_g to values of the order of τ.

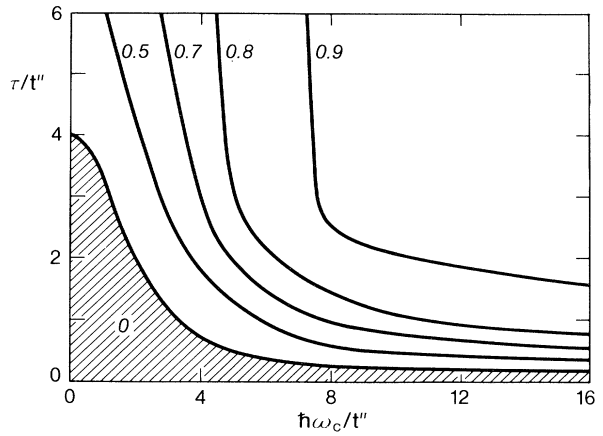

FIG. 2. The values of Δ_g/τ as a function of magnetic field $\hbar\omega_c/t''$ and τ/t''' for $t'/t=0.2$ and for $t''\ll t'$. The shaded region corresponds to $\Delta_g=0$ and band overlap, the semimetallic case. The gap increases rapidly to its maximum value of $\Delta_g=\tau$.

at $\alpha=\alpha_c$, to a value of the order of 1 for $\alpha\gg 1$. (C) For $\tau/t''>4$ there is a gap Δ_g for $\alpha=0$ [see Eq. (3)], which increases monotonically as α increases.

Because of the intricate fine structure of the spectrum, characteristic of Bloch electrons in a magnetic field, small gaps could appear at the Fermi level even for $\alpha<\alpha_c$ for some particular values of the magnetic field. These are, in general, smaller gaps which disappear as the field either increases or decreases in strength, i.e., they form "islands" of small-gap states in general areas of semimetallicity. For discussion purposes we use in what follows the terms semiconductor and semimetal in a qualitative way: we refer specifically to the situation $\alpha>\alpha_c$ and $\alpha<\alpha_c$, respectively.

In Fig. 2 we present a diagram of constant energy-gap parameter Δ_g/τ in the $(\alpha,\tau/t'')$ plane. For $\tau/t''<4$ there is a two-dimensional region of the plane corresponding to the semimetallic regime, $\Delta_g=0$. The high-magnetic-field region is semiconducting, with the energy-gap parameter increasing monotonically along both axes.

A few other points deserve comment:

(i) The order parameter τ must be calculated self-consistently, and in principle should depend on the direction and strength of the magnetic field. The equilibrium value of τ is given by

$$\tau=\lambda\sum_{nk_\perp\sigma}\cos(nQ_x a_1)\langle c^\dagger_{nk_\perp\sigma}c_{n(k_\perp+Q_\perp)\sigma}\rangle,$$

where λ is an interaction parameter, and $\langle\cdots\rangle$ indicates the thermodynamic average. According to the densities of states calculated for high magnetic fields, we expect τ to increase with field up to values of the order of those obtained for a perfectly nesting Fermi surface. We thus expect the effect of the magnetic field to be even more pronounced.

(ii) Because of the nature of the electronic states at the Fermi level, which in the absence of a DW form a continuum even for finite magnetic fields,[11] we do not expect the thermodynamics of the system to change appreciably. If the DW critical temperature at zero magnetic field is not too low, it will not be sensitive to the application of any reasonable magnetic field, especially if lifetime effects play any role.

(iii) As pointed out in Ref. 8, for some particular values of the magnetic fields and the band parameters, the system could lower its energy by shifting the **Q** vector, i.e., by changing the periodicity of the DW. This magnetic-field–induced transition may occur in the semimetallic regime. In the case of a CDW other contributions to the total energy, which tend to lock in the **Q** vector, may however oppose this transition.[17,18]

(iv) The results reported here are not a consequence of a half-filled band. Similar results appear (albeit with a different **Q** vector) for other electron concentrations.

In conclusion, we have shown that a high magnetic field may induce a metal-insulator transition in Q1D systems with a DW arising from an imperfectly nested Fermi surface. Our theory provides a plausible explanation for the anomalies observed in $NbSe_3$ at high fields. Because of the large number of atoms in the primitive unit cell, the Fermi surface of $NbSe_3$ consists of several corrugated surfaces, each one generally speaking associated with one chain and one band. According to band-structure calculations,[19] the normal-state Fermi surface consists of several open sheets

(corrugated planes), and an inner multiply connected surface. At the low-temperature transition T_2, the CDW removes parts of one of the open sheets, probably leaving some electron and hole small pockets. A high magnetic field may shrink considerably or even completely obliterate these small pieces. The residual conductivity[4,5] and de Haas–Shubnikov oscillations[20,21] at very high fields may then be associated with either the inner, multiply connected Fermi surface, and/or with the leftover parts of the open surface, which must exist before a real gap opens up at high fields.

The effect described here does not occur for fields perpendicular to the "corrugated plane" of the Fermi surface, in agreement once again with the experimental results.[5]

We are deeply indebted to Professor R. V. Coleman for many illuminating and challenging discussions, and for keeping us informed on a day-to-day basis of the results of his experiments. This work was supported by the National Science Foundation through Grant No. DMR81-06494. One of us (C.A.B.) would like to acknowledge the financial support of a J. S. Guggenheim Memorial Foundation Fellowship and of the Centro Atómico Bariloche.

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[16]Because of the mixing of $k_\perp$ and $k_\perp + Q_\perp$, it is convenient to define a 2×2 matrix Green's function. The continuous-fraction method used here is a generalization of the one described in Ref. 11 to 2×2 matrices instead of scalars.

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