Excitonic Insulator in a Magnetic Field

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The effects of large magnetic fields on the excitonic insulator are considered. Since the excitonic insulator does not display a Meissner effect (according to a recent paper by Jerome, Rice, and Kohn), a Hartree-Fock ground state can be expressed in terms of Landau-level basis functions. A magnetic-field-dependent phase diagram and the magnetoconductivity are discussed. The excitonic insulator, not yet observed experimentally, may be observable in high magnetic fields at normal pressure.

I. INTRODUCTION

THE excitonic insulator has been discussed in a number of recent papers.[1–12] This new phase, not yet observed experimentally, is expected to occur in solids with bandgap or band overlap less than the exciton binding energy. Below a critical temperature, the single-particle energy spectrum contains a gap similar to the energy gap in a superconductor. The ground state is a coherent state of excitons rather than the filled valence band, empty conduction band of a normal insulator. Diagonal long-range order exists in the electron assembly, characterized by the wave vectors in the Brillouin zone which separate valence band maxima from conduction band minima (of the normal phase). Collective excitations obey an acoustic-mode dispersion relation.[6]

Recently, the phase transition characteristics have been discussed by Kohn.[11,12] The preliminary results indicate that passing from low-density to high-density limits at zero temperature involves an infinite sequence of second-order phase transitions and excitonic phases.

We consider the excitonic insulator in arbitrary magnetic field. Although it has been suggested that for triplet exciton states the excitonic insulator will display antiferromagnetic ordering,[4,13,14] at the present time we will consider only a spinless system. The total charge of the basic group is zero, which precludes the Meissner effect according to arguments due to Yang.[15] (A direct proof occurs in Ref. 6 as well.) Therefore the excitonic insulator in a magnetic field can be conveniently described in terms of Landau-level basis functions.

Two principal effects occur when a magnetic field is applied:

(1) The bandgap is replaced by an effective bandgap which has been increased by the cyclotron energies of $n=0$ Landau levels in valence and conduction bands[16] (in the absence of spin). The phase diagram is no longer centered about $G=0$, but occurs near $G+\frac{1}{2}\hbar\omega_{\mu c}=0$. G is the bandgap (or band overlap for negative values) and μ indicates the reduced mass.

(2) The exciton binding energy increases with magnetic field.[17,18] Representing the magnetic-field-dependent binding energy by $E_B(H)$, the excitonic insulator regime $-E_B(H)\lesssim G+\frac{1}{2}\hbar\omega_{\mu c}\leq E_B(H)$ expands as the magnetic field increases.

In the following sections we will discuss the magnetic-field-dependent ground state, phase diagram, and electrical conductivity. Possible observation of the excitonic insulator phase in high magnetic fields at normal pressure will be considered, in contrast to the high-pressure experiments required when the field is zero.

II. GROUND STATE

In this section, we will discuss the ground state in terms of the stationary states of spinless single particles in a magnetic field, using a BCS type of variational calculation.[19] Consider spinless particles with single valence and conduction bands which have isotropic and quadratic E versus $\mathbf{k}$ relations, and in which the band extrema are separated by a wave vector $\mathbf{w}$ in the Brillouin zone. Choosing the field $\mathbf{H}$ in the z direction and the gauge $\mathbf{A}_H=(-yH,0,0)$, the single-particle wave functions are

$$\phi_\alpha(\mathbf{x})\equiv|n,k_x,k_z\rangle=e^{ik_xx+ik_zz}\chi_n[y+(k_x/m_j\omega_{jc})], \quad (1)$$

where j refers to band a or band b, and the crystal has unit dimensions. The χ_n are normalized one-dimensional

[1] J. des Cloizeaux, J. Phys. Chem. Solids **26**, 259 (1965).
[2] L. V. Keldysh and Yu. V. Kopaev, Fiz. Tverd. Tela **6**, 2791 (1964) [English transl.: Soviet Phys.—Solid State **6**, 2219 (1965)].
[3] A. N. Kozlov and L. A. Maksimov, Zh. Eksperim. i Teor. Fiz. **48**, 1184 (1965) [English transl.: Soviet Phys.—JETP **21**, 790 (1965)].
[4] A. N. Kozlov and L. A. Maksimov, Zh. Eksperim. i Teor. Fiz. **49**, 1284 (1965) [English transl.: Soviet Phys.—JETP **22**, 889 (1965)].
[5] Yu. V. Kopaev, Fiz. Tverd. Tela **8**, 223 (1966) [English transl.: Soviet Phys.—Solid State **8**, 175 (1966)].
[6] D. Jerome, T. M. Rice, and W. Kohn, Phys. Rev. **158**, 462 (1967).
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[15] C. N. Yang, Rev. Mod. Phys. **34**, 694 (1962).
[16] I. P. Batra and R. R. Haering, Can. J. Phys. **45**, 3401 (1967).
[17] Y. Yafet, R. W. Keyes, and E. N. Adams, J. Phys. Chem. Solids **10**, 254 (1956).
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[19] J. Bardeen, L. N. Cooper, and J. R. Schrieffer, Phys. Rev. **108**, 1175 (1957).

harmonic oscillator functions. Figure 1 shows the magnetic-field-dependent band structure for reference.

Consider the operators

$$\psi_a(\mathbf{x})=\sum_\alpha a_\alpha\phi_\alpha(\mathbf{x})$$

and

$$\psi_b(\mathbf{x})=\sum_\alpha b_\alpha\phi_\alpha(\mathbf{x}). \tag{2}$$

a_α and b_α are fermion destruction operators for Landau-level states in bands a and b. Summations are over the Brillouin zone. We take for a model Hamiltonian

$$\mathcal{H}=\sum_{j=a,b}\int\psi_j^\dagger(\mathbf{x})\epsilon_j(\mathbf{x})\psi_j(\mathbf{x})d\mathbf{x} +\frac{1}{2}\int\rho(\mathbf{x})\rho(\mathbf{x}')V(\mathbf{x}-\mathbf{x}')d\mathbf{x}d\mathbf{x}', \tag{3}$$

where

$$\epsilon_a(\mathbf{x})=-\tfrac{1}{2}G+(2m_a)^{-1}(\nabla+ie\mathbf{A}_H)^2,$$
$$\epsilon_b(\mathbf{x})=\tfrac{1}{2}G-(2m_b)^{-1}(\nabla+ie\mathbf{A}_H)^2, \tag{4}$$
$$(\hbar=c=1),$$

and the charge density is represented by

$$\rho(\mathbf{x})=\psi_a^\dagger(\mathbf{x})\psi_a(\mathbf{x})+\psi_b^\dagger(\mathbf{x})\psi_b(\mathbf{x}). \tag{5}$$

$V(\mathbf{x}-\mathbf{x}')$ is the Coulomb interaction screened by an effective dielectric constant.

We introduce a Hartree-Fock trial wave function for the ground state

$$|\Psi\rangle=\prod_\alpha c_\alpha^*|\text{vac}\rangle. \tag{6}$$

c_α^* creates an electron in a linear normalized combination of band a and band b Landau-level states:

$$c_\alpha=u_\alpha a_\alpha-v_\alpha b_\alpha. \tag{7}$$

We obtain a trial value for the energy

$$W=\langle\Psi|\mathcal{H}|\Psi\rangle=\sum_\alpha\epsilon_a(\alpha)u_\alpha^2+\sum_\alpha\epsilon_b(\alpha)v_\alpha^2 +\sum_{\alpha,\alpha'}V_{\alpha\alpha'}u_\alpha v_\alpha u_{\alpha'}v_{\alpha'}, \tag{8}$$

where

$$V_{\alpha\alpha'}=\int V(|\mathbf{x}-\mathbf{x}'|)\phi_\alpha^*(\mathbf{x})\phi_\alpha(\mathbf{x}') \times\phi_{\alpha'}^*(\mathbf{x}')\phi_{\alpha'}(\mathbf{x})d\mathbf{x}d\mathbf{x}'. \tag{9}$$

The single-particle energies are

$$\epsilon_a(\alpha)\equiv\epsilon_a(n_a,k_{az})=-\tfrac{1}{2}G-(n_a+\tfrac{1}{2})\omega_{ac}-(2m_a)^{-1}k_{az}^2,$$
$$\epsilon_b(\alpha)=\tfrac{1}{2}G+(n_b+\tfrac{1}{2})\omega_{bc}+(2m_b)^{-1}k_{bz}^2, \tag{10}$$

where k_{ax}, k_{az} etc., are measured with respect to the band extrema. Intraband electron-electron interactions do not appear in (8) in this simple model due to the exclusion principle. (Single-particle terms are not completely analogous to the corresponding terms obtained by Des Cloiseaux[1] and by Jerome, Rice, and Kohn (JRK),[6] as they did not include all single-particle kinetic terms from the valence band. These terms do not contribute to subsequent calculations in any case.)

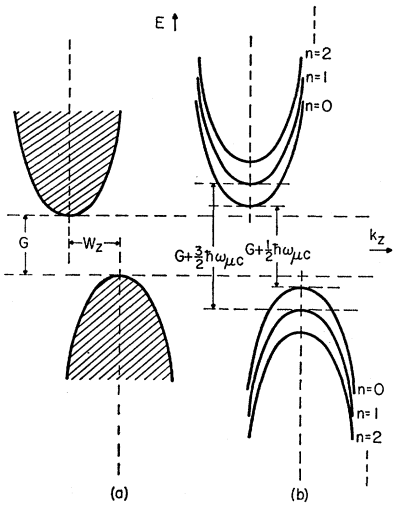

FIG. 1. (a) Band structure in the absence of a magnetic field. (b) Band structure in a magnetic field, but without regard to intrinsic spin.

Minimizing W, we obtain

$$u_\alpha=[\tfrac{1}{2}(1+\epsilon_\alpha/E_\alpha)]^{1/2}, \tag{11}$$

$$v_\alpha=[\tfrac{1}{2}(1-\epsilon_\alpha/E_\alpha)]^{1/2}, \tag{12}$$

where

$$\epsilon_\alpha=\tfrac{1}{2}[\epsilon_b(\alpha)-\epsilon_a(\alpha)] \tag{13}$$

and

$$E_\alpha^2=\epsilon_\alpha^2+\Delta_\alpha^2. \tag{14}$$

We have chosen the phase of Δ_α, which is arbitrary, as zero. The gap function satisfies

$$\Delta_\alpha=\sum_{\alpha'}V_{\alpha\alpha'}\frac{\Delta_{\alpha'}}{2E_{\alpha'}}. \tag{15}$$

Equation (15) may be written as

$$\left\{\left[G+(n+\tfrac{1}{2})\omega_{\mu c}+\frac{k_z^2}{2\mu}\right]^2+4\Delta_\alpha^2\right\}^{1/2}\xi(\alpha) =\sum_{\alpha'}V_{\alpha\alpha'}\xi(\alpha'), \tag{16}$$

where

$$\xi(\alpha)=\Delta_\alpha/2E_\alpha. \tag{17}$$

III. PHASE DIAGRAM

The wave equation for the exciton[20] in the Landau representation is obtained by expanding

$$\zeta(\mathbf{r}_1\mathbf{r}_2)=\sum_\alpha\zeta(\alpha)\phi_\alpha^*(\mathbf{r}_1)\phi_\alpha(\mathbf{r}_2)$$

[20] R. S. Knox, in *Solid State Physics*, edited by F. Seitz and D. Turnbull (Academic Press Inc., New York, 1963), Suppl. 5.

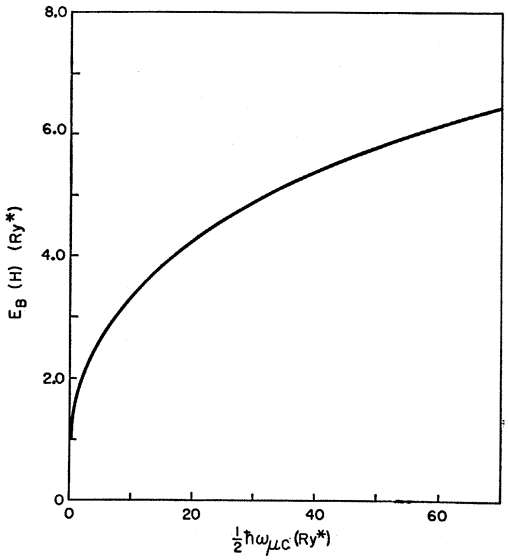

FIG. 2. Binding energy of the exciton as a function of magnetic field, after Yafet, Keyes, and Adams (see Ref. 17).

and the result is

$$\left[(n+\tfrac{1}{2})\omega_{\mu c}+\frac{k_z^2}{2\mu}+(|E_B(H)|-\tfrac{1}{2}\omega_{\mu c})\right]\zeta(\alpha)$$
$$=\sum_{\alpha'} V_{\alpha\alpha'}\zeta(\alpha'). \quad (18)$$

Comparing (18) with (16), $\Delta=0$ when $G\geq E_B(H)-\frac{1}{2}\hbar\omega_{\mu c}$. Δ has a maximum value at $G+\frac{1}{2}\hbar\omega_{\mu c}=0$, and the phase diagram for $G+\frac{1}{2}\hbar\omega_{\mu c}\geq 0$ is identical in form to the zero-field diagram calculated by Kozlov and Maksimov for $G\geq 0$.[3]

In the semimetallic band-overlap regime, the phase boundary is determined by changes in the dielectric function which reduce the effective interaction $V(\mathbf{x}-\mathbf{x}')$ as band overlap increases. When the magnetic field is absent, the effective dielectric constant is approximately

$$K(q)=\frac{K_0}{q^2+1/\lambda^2}, \quad (19)$$

as taken by Kozlov and Maksimov. K_0 is the static dielectric constant and λ is the Thomas-Fermi screening length.[21] Although we can assume that K_0 is only slightly changed by an applied magnetic field, the complete magnetic-field-dependent dielectric function is considerably more complex than (19).[22–24] We will not discuss this question in detail in regard to the excitonic insulator, but will only comment that we expect the gross characteristics of the $G+\frac{1}{2}\hbar\omega_{\mu c}<0$ part of the phase diagram to be similar to the $G<0$ regime at zero field, although the extent of this regime may be reduced at high fields.

The exciton binding energy as a function of magnetic field is shown in Fig. 2.[17,18] The zero-field phase diagram of Ref. 3, Δ versus G, is scaled on both axes according to $E_B(H)$ when a magnetic field is applied. The scaled diagram is shifted on the G axis by $-\frac{1}{2}\hbar\omega_{\mu c}$. At $G+\frac{1}{2}\hbar\omega_{\mu c}=0$, the gap function maximum is $\Delta_{\max}=(8/\pi^2)E_B(H)$. Figure 3 illustrates the magnetic-field-dependent phase diagram.

IV. MAGNETOCONDUCTIVITY

We will consider the longitudinal dc conductivity using the general response theory derived by JRK.[6] The response of the excitonic insulator to an electric field perpendicular to H is considerably more difficult to calculate since in this case off-diagonal elements of the Green's functions must be determined. (However, the cyclotron resonance can be determined in a straightforward manner).

From the equations of motion of the Heisenberg operators, diagonal elements of interband and intraband Green's functions in the Landau basis are, at zero temperature,[6]

$$G_b(\alpha,\epsilon)=\frac{\epsilon-\epsilon_a(\alpha)}{[\epsilon-\epsilon_a(\alpha)][\epsilon-\epsilon_b(\alpha)]-\Delta_\alpha{}^2}, \quad (20)$$

$$F^\dagger(\alpha,\epsilon)=\frac{\Delta_\alpha{}^\dagger}{[\epsilon-\epsilon_a(\alpha)][\epsilon-\epsilon_b(\alpha)]-\Delta_\alpha{}^2}, \quad (21)$$

where

$$G_b(1,1')=-i\langle T\psi_b(1)\psi_b{}^\dagger(1')\rangle$$
$$=\sum_{\alpha,\epsilon} G_b(\alpha,\epsilon)\phi_\alpha{}^*(\mathbf{x}_1)\phi_\alpha(\mathbf{x}_1')\exp[i\epsilon(t_1'-t_1)] \quad (22)$$

and

$$G_a(1,1')=-i\langle T\psi_a(1)\psi_a{}^\dagger(1')\rangle, \quad (23)$$

$$F^\dagger(1,1')=-i\langle T\psi_a(1)\psi_b{}^\dagger(1')\rangle, \quad (24)$$

$$F(1,1')=-i\langle T\psi_b(1)\psi_a{}^\dagger(1')\rangle. \quad (25)$$

T is the time-ordering operator and the variable (1) in the Heisenberg operator denotes $(\mathbf{x}_1,t_1)$. Expressions analogous to (20) and (21) are obtained for $G_a(\alpha,\epsilon)$ and $F(\alpha,\epsilon)$.

The gap function is

$$\Delta_\alpha{}^\dagger=i\sum_{\alpha'}\sum_{\epsilon'} V_{\alpha\alpha'}F^\dagger(\alpha',\epsilon') \quad (26)$$

and Eq. (15) is recovered, using (21) in (26).

We introduce a vector potential $\mathbf{A}(\mathbf{x},t)$ as a perturbation to the Hamiltonian of Eq. (3), representing a weak electric field. With the perturbation the single-

[21] J. M. Ziman, *Electrons and Phonons* (Clarendon Press, Oxford, England, 1960).
[22] E. N. Adams and T. D. Holstein, J. Phys. Chem. Solids **10**, 254 (1959).
[23] P. N. Argyres, Phys. Rev. **148**, 548 (1966).
[24] O. Wolman and A. Ron, Phys. Rev. **148**, 548 (1966).

particle terms become

$$\epsilon_j'(1,1')=\epsilon_j(1)-\frac{e}{m_j}[i\nabla_1-e\mathbf{A}_H(1)]\cdot\mathbf{A}(1)\delta(1,1') -v(1-\bar{1})\langle\rho(\bar{1};\mathbf{A}_H,\mathbf{A})\rangle\delta(1,1'), \quad (27)$$

where $j=a, b$. In the last term, ρ is the density matrix and v the unscreened Coulomb potential. The bar over the variable denotes integration.

The diamagnetic current response is the usual expression

$$\mathbf{J}^d(1)=-(n_c e^2/\mu)\mathbf{A}(1), \quad (28)$$

where n_c is the number of carriers in either band. (Apart from the small Landau diamagnetism, diamagnetic and paramagnetic currents due to $\mathbf{A}_H$ cancel.[6])

The paramagnetic response is given by

$$\mathbf{J}^p(1)=-\tfrac{1}{2}e\left\{[\nabla_1+ie\mathbf{A}_H(1)-\nabla_{1'}+ie\mathbf{A}_H(1')]\times\left[m_b^{-1}\frac{\delta G_b(1,1')}{\delta\mathbf{A}(\bar{2})}-m_a^{-1}\frac{\delta G_a(1,1')}{\delta\mathbf{A}(\bar{2})}\right]\right\}_{1'=1}\cdot\mathbf{A}(\bar{2}) =\mathbf{J}_b{}^p(1)+\mathbf{J}_a{}^p(1). \quad (29)$$

$\delta G(1,1')/\delta\mathbf{A}(2)$ denotes the functional derivative, which can be obtained from $\delta\epsilon_j'(1,1')/\delta\mathbf{A}(2)$ by using the general response equations derived by JRK. Since the equations are lengthy, they will not be repeated here, and the reader is referred to Eqs. (3.27)–(3.31) of Ref. 6.

$$\frac{\delta\epsilon_j'(1,1')}{\delta\mathbf{A}(2)}=\frac{-e}{2m_j i}\{[\nabla_2+ie\mathbf{A}_H(2)-\nabla_{2'}+ie\mathbf{A}_H(2')]\times\delta(1-2)\delta(1'-2')\}_{2'=2} -v(1-\bar{1})[\delta\langle\rho(\bar{1})\rangle/\delta\mathbf{A}(2)]\delta(1',1). \quad (30)$$

Landau transforming the response equations, we are led to

$$\frac{\delta G_b(1,1')}{\delta A(2)_z}=\frac{-e}{2m_b}\sum_{\substack{\alpha,\alpha'\\ \epsilon,\epsilon'}}(k_z'+k_z)[G_b(\alpha',\epsilon')G_b(\alpha,\epsilon) -F(\alpha',\epsilon')F^\dagger(\alpha,\epsilon)]\exp\{it_2(\epsilon-\epsilon')+it_1(\epsilon'-\epsilon)\} \times\phi_\alpha{}^*(\mathbf{x}_1)\phi_\alpha(\mathbf{x}_2)\phi_{\alpha'}{}^*(\mathbf{x}_2)\phi_{\alpha'}(\mathbf{x}_1) +(\text{terms in } v_{\alpha\alpha'} \text{ and } V_{\alpha\alpha'}). \quad (31)$$

The interaction terms are not expressed explicitly since we will be confining the discussion to the equal mass, semimetallic limit in which all such terms eventually cancel by particle hole symmetry.[6] Henceforth they will be dropped.

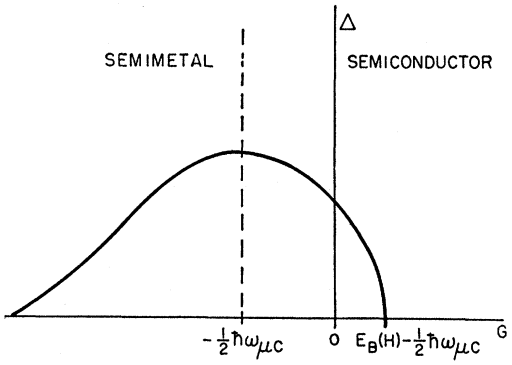

FIG. 3. Magnetic-field-dependent phase diagram of the excitonic insulator, ignoring spin.

Applying the current operator of Eq. (29), we obtain

$$J_b{}^p(1)_z=\frac{-ie^2}{2m^2}\sum_{\substack{\alpha,\alpha'\\ \epsilon\\ \omega,q}}(2k_z+q_z)^2[G_b(\alpha',\epsilon+\omega)G_b(\alpha,\epsilon) -F(\alpha',\epsilon+\omega)F^\dagger(\alpha,\epsilon)]\exp\{i\omega(t_1-t_2)\} \times\phi_\alpha{}^*(\mathbf{x}_1)\phi_{\alpha'}(\mathbf{x}_1)A_{\alpha\alpha'}(q,\omega)_z, \quad (32)$$

where

$$A_{\alpha\alpha'}(q,\omega)_z=A(q,\omega)_z\int\phi_{\alpha'}{}^*(\mathbf{x}_2)e^{i\mathbf{q}\cdot\mathbf{x}_2}\phi_\alpha(\mathbf{x}_2)d\mathbf{x}_2 \quad (33)$$

and

$$|\alpha'\rangle\equiv|n',\ k_x+q_x,\ k_z+q_z\rangle, \quad |\alpha\rangle\equiv|n,k_x,k_z\rangle. \quad (34)$$

We will be concerned with the limit $q\to 0$ and near that limit the $A_{\alpha\alpha'}(q,\omega)$ are vanishingly small unless $n'=n$. Considering only the $n'=n$ terms in (32), and noting that the Green's functions do not depend on k_x or k_x', the summation over k_x, $(q=0^+)$, yields

$$J_b{}^p(0^+,\omega)_z=\frac{-ie^2\omega_c}{2\pi m}\sum_{\substack{n,k_z\\ \epsilon}}(2k_z+q_z)^2[G_b(n,\ k_z+q_z;\ \epsilon+\omega) \times G_b(n,k_z,\epsilon)-F(n,\ k_z+q_z;\ \epsilon+\omega) \times F^\dagger(n,k_z;\ \epsilon)]A(q,\omega)_z|_{q=0^+}, \quad (35)$$

where we have used

$$\lim_{q\to 0}\sum_{k_x}\chi_n{}^*\left(y+\frac{k_x+q_x}{m\omega_c}\right)\chi_n\left(y+\frac{k_x}{m\omega_c}\right)=\frac{m\omega_c}{2\pi}, \quad (36)$$

valid when the cyclotron radius is much smaller than the dimensions of the crystal.

We proceed directly to the finite temperature generalization of (35) by replacing the functions G, F with the temperature-dependent Green's functions

$$\mathcal{G}_j(1,1')=-i\langle\langle T\psi_j(1)\psi_j{}^\dagger(1')\rangle\rangle, \quad \mathcal{F}^\dagger(1,1')=-i\langle\langle T\psi_a(1)\psi_b{}^\dagger(1')\rangle\rangle, \quad \mathcal{F}(1,1')=-i\langle\langle T\psi_b(1)\psi_a{}^\dagger(1')\rangle\rangle. \quad (37)$$

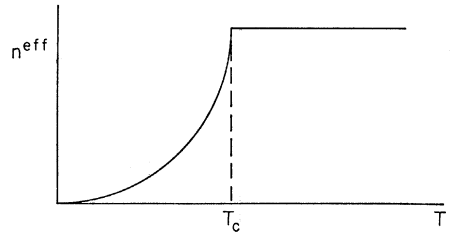

FIG. 4. n^{eff} versus temperature. The dc conductivity of the excitonic insulator in a large, longitudinal magnetic field is $n^{\text{eff}}(H,T)\sigma_0(H,T)/2n_c$, where $\sigma_0(H,T)$ is the temperature- and field-dependent conductivity of the normal phase. The field dependence of n^{eff} enters through the dependence of Δ on $E_B(H)$.

These functions are obtained from (20) to (25) using the fact that the real parts do not depend on temperature and the imaginary and real parts are related by a well-known dispersion equation.[25] The resulting functions are identical in form to the zero-field functions obtained by JRK, Eqs. (6.3) of Ref. 6. The current density is obtained using these expressions for $\mathcal{G}$ and $\mathcal{F}$ in (35) and the corresponding equation for $J_a{}^p(0^+,\omega)_z$.

$$\lim_{\omega\to 0} J^p(0,\omega)_z = \frac{e^2\omega_c}{2\pi m}\sum_{n,k_z}\frac{k_z{}^2\Delta^2}{E_{n,k_z}{}^3} \times[1-2f(n,k_z)]U(0,\omega)_z|_{\omega\to 0}, \quad (38)$$

where $f(\alpha)\equiv f(n,k_z)=[\exp(E_\alpha/kT)+1]^{-1}$ is the Fermi distribution function, and E_α is specified by Eq. (14).

The diamagnetic contribution for the equal-mass semimetallic case is

$$J^d(0,0)_z = \frac{-e^2\omega_c}{2\pi}\sum_{n,k_z} 2v_\alpha{}^2 A(0,0)_z = \frac{-e^2\omega_c}{2\pi}\sum_{n,k_z}(1-\epsilon_\alpha/E_\alpha)A(0,0)_z \quad (39)$$

using (12) in the second equation. It can be shown by integrating that the diamagnetic current cancels the temperature-independent term of (38), and the total current is

$$J(0,0)_z = \frac{-e^2\omega_c}{\pi m}\sum_{n,k_z}\frac{k_z{}^2\Delta^2}{E_{n,k_z}{}^3}f(n,k_z)A(0,0)_z. \quad (40)$$

The vector potential is

$$A(0,\omega)_z = \frac{-i}{\omega}E_z(\omega) \quad (41)$$

and thus the low-frequency conductivity is

$$\sigma_{zz}(0,\omega) = \frac{ie^2\omega_c}{\pi m\omega}\sum_{n,k_z}\frac{k_z{}^2\Delta^2}{E_{n,k_z}{}^3}f(n,k_z) = \frac{ie^2 n^{\text{eff}}}{m\omega}, \quad (42)$$

where

$$n^{\text{eff}} = \frac{m\omega_c}{\pi}\sum_{n,k_z}\frac{k_z{}^2\Delta^2}{mE_{n,k_z}{}^3}[\exp(E_{n,k_z}/kT)+1]^{-1}. \quad (43)$$

In the materials of greatest interest such as the semimetals, in the excitonic insulator regime near $G+\frac{1}{2}\hbar\omega_{\mu c}=0$ the effective range of band overlap will include only the $n=0$ Landau level, all other levels falling completely above or below. Using this fact, and also the number of free carriers in the extreme quantum limit, in either band,

$$n_c = \sum_{k_z=}^{k_z(E_f)}\frac{m\omega_c}{2\pi} = \frac{m\omega_c}{2\pi}k_z(E_f), \quad (44)$$

an expression for n^{eff} is obtained,

$$n^{\text{eff}} = 2n_c\int_0^\infty \frac{d\epsilon_\alpha\Delta^2}{(\epsilon_\alpha{}^2+\Delta^2)^{3/2}}\left[\exp\left(\frac{\epsilon_\alpha{}^2+\Delta^2}{k^2T^2}\right)^{1/2}+1\right]^{-1} = 2n_c\int_0^\infty \frac{d\nu}{(\nu^2+1)^{3/2}}\{\exp[\Delta(\nu^2+1)/kT]+1\}^{-1}. \quad (45)$$

Equation (45) is identical in form to the zero-field expression obtained by JRK. When $\Delta=0$ at $T\geq T_c$, where T_c is the critical temperature, $n^{\text{eff}}=2n_c$. When $\Delta\neq 0$ and $T\to 0$, $n^{\text{eff}}=0$. n^{eff} is depicted schematically in Fig. 4.

An expression for the real part of the conductivity can be obtained by introducing a finite level width $\Gamma=(1/\tau)$, where τ is some average lifetime. We let $\omega\to\omega+i/\tau$ in Eq. (42), and obtain for the real part of the dc conductivity,

$$\text{Re}(\sigma_{zz}) = n^{\text{eff}}e^2\tau/m. \quad (46)$$

In (46) we have taken τ, a function of magnetic field and electron energy, outside of the integral in n^{eff}. Since the integrand is sharply peaked at the Fermi energy, this procedure may be a reasonable approximation, with τ referring to an average lifetime at the Fermi surface (in the presence of a magnetic field).

V. DISCUSSION

It must first be emphasized that the quantum limit for a semimetal with free carriers due to band overlap is not similar to the quantum limit for a metal. In a metal, the Fermi energy increases when the $n=0$ Landau level exceeds the zero-field Fermi level. In the absence of intrinsic spin, we expect the contrary

[25] L. D. Landau, Zh. Eksperim. i Teor. Fiz. 34, 262 (1958) [English transl.: Soviet Phys.—JETP 7, 182 (1958)].

behavior in a semimetal, with a decrease of charge carrier density to zero ($T=0$) when the $n=0$ Landau levels of valence and conduction bands fail to overlap. At higher magnetic fields a decrease of the Fermi level will occur as the $n=0$ level of the valence band moves downward from the band maximum.

In our calculations we have ignored intrinsic spin, and we will now show that no complications arise from this source. For the Wannier excitons of this theory, appropriate to materials in which $|G|$ is small, spin-orbit coupling is practically identical in bound and free states, and intrinsic spin does not contribute a term to the binding energy $E_B(H)$. However the effective band gap becomes $G^*=G+\frac{1}{2}\hbar\omega_{\mu c}\pm\frac{1}{2}\hbar\omega_{es}\pm\frac{1}{2}\hbar\omega_{hs}$, where $\hbar\omega_{es}$ and $\hbar\omega_{hs}$ refer to spin splitting of $n=0$ electron and hole Landau levels. Because of screening by quasifree electrons, we are concerned only with the regime corresponding to the minus signs. (Excitonic insulator regimes which might be expected to occur for the other cases are "screened out.")

The effective band gap G^* will increase or decrease with increasing magnetic field if spin splitting is respectively smaller or greater than the Landau-level spacing. The excitonic insulator phase may be introduced in either semimetals or small bandgap semiconductors by sufficiently large magnetic fields (or be removed, if occurring at zero field in some complex and poorly understood substance, such as an organic compound.)

Zittartz has considered the effects of impurity scattering on the excitonic insulator,[8–10] and obtained an impurity-dependent critical temperature

$$kT_c=kT_{c0}-\pi\hbar/4\tau;\quad kT_c>0 \tag{47}$$

and

$$\tau(T_c=0)\sim\hbar/kT_{c0}.$$

T_{c0} is the critical temperature in the absence of impurities and τ is the relaxation time. [Equation (47) can be justified by a simple uncertainty principle argument.] With T_{c0} the order of one degree Kelvin, and a Fermi velocity $\lesssim 10^6$ cm/sec, the critical mean free path for $kT_c\neq 0$ is $\lesssim 10^{-5}$ cm. Contrary to the discussion in Ref. 8, this is not an experimental obstacle as mean free paths the order of 1 cm have been obtained in bismuth[26] and several other metals (Mg, Sn, Ga, Tl). In this case, Eq. (46) is representative.

The scale of the phase diagram of Fig. 3 depends on the binding energy of the exciton in a magnetic field, Fig. 2, and position along the G axis is determined by the cyclotron energy (and also the spin splitting). Variation of $E_B(H)$ and the ratio $\frac{1}{2}\hbar\omega_{\mu c}/G$ are greatest in materials which have low effective mass, high dielectric constant, and small bandgap or band overlap. The semimetals arsenic, antimony, and bismuth are favorable in this regard, and we consider bismuth where $\mu\sim 0.01m_e$[27] and $K_0\sim 100$.[28] The ratio of cyclotron energy to the effective rydberg is proportional to $(m_eK_0/\mu)^2$, and this factor may be as much as 10^8 greater in bismuth than in the hydrogen atom. At 50 kG, $\frac{1}{2}\hbar\omega_{\mu c}\sim 2500$ Ry*. Using Fig. 2, and the fact that at high fields the exciton binding energy is approximately proportional to $H^{1/3}$, we find E_B (50 kG)$\sim$20 Ry*. (For bismuth, Ry*$\sim 1.4\times 10^{-5}$ eV.) The total number of electrons and holes will be greater than 10^{14} cm^{-3} in the new phase. The transition temperature is order unity times Δ or $E_B(H)$, and we find T_c(50 kG)$\sim$3°K for bismuth, which compares with $\sim$0.15°K when the magnetic field is zero. E_B(50 kG) corresponds to a magnetic field range $\delta H\sim 400$ G.

The phase diagram migrates along the bandgap axis toward increasing or decreasing band overlap when spin splitting is less than or greater than the Landau-level spacing. (In Fig. 2 we have taken the spin splitting as zero for simplicity.) At 50 kG the cyclotron energy in bismuth is as great as $\sim$0.03 eV. The zero-field Fermi energy (band overlap) is $\sim$0.02 eV, and we can expect that the excitonic insulator in the extreme quantum limit will be attainable experimentally in materials such as bismuth. The ratio of spin splitting to Landau-level spacing in the semimetals is not yet well understood, however, in bismuth the spin splitting is known to exceed the Landau-level spacing considerably with H along the trigonal axis. (If spin splitting and Landau-level spacing are nearly equal, the effective band gap is relatively insensitive to magnetic field.) Recent experiments show that in a $Bi_{1-x}Sb_x$ alloy with "x" sufficiently great to remove band overlap, band overlap can be reintroduced by applying a large magnetic field.[29] (This alloy is somewhat unusual in that mean free paths of free carriers remain long even when x is several atomic percent.)

Experimental circumstances may favor observation of the excitonic insulator phase in high magnetic fields rather than at high pressures. Pressure-control requirements are probably more exacting than present experimental techniques permit. However the lower symmetry in a magnetic field may introduce extraneous and unwelcome effects which remove the excitonic insulator regime, and which are difficult to evaluate *a priori*. Experiments to observe the excitonic insulator at high magnetic fields in the $Bi_{1-x}Sb_x$ alloy are being conducted at the National Research Council of Canada.

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