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[5]Free-atom properties were calculated using a spin-polarized Kohn-Sham program written by J. F. Janak (private communication). The free-atom electron density was made spherically symmetric by partially filling the discrete-atom levels where necessary.

[6]The calculations used for Fig. 3 are self-consistent at every distance; only one of these distances minimizes the total energy of a given system. [The calculated atomic binding energies (cf. Refs. 1 and 5) are 1.3 eV (Li), 3.0 eV (Si), and 3.6 eV (Cl).]

[7]K. Y. Yu, J. N. Miller, P. Chye, W. E. Spicer, N. D. Lang, and A. R. Williams, Phys. Rev. B (to be published).

[8]This calculation used first-order perturbation theory. The magnitude of the first-order terms suggests that second-order terms can be important (Ref. 7). For adsorption in a threefold site on Al(111), the first-order treatment gives physically reasonable results for Si ($d = 1.6$ a.u., corresponding to an Al-Si bond length $b = 2.6$ Å) and Li ($d = 2.1$ a.u., corresponding to $b = 2.8$ Å), but indicates that the Cl atom sits further out than in the jellium calculation, which is presumably not the case. The use of first-order perturbation theory in this context cannot therefore be considered to represent a generally adequate treatment of discrete-lattice effects.

[9]Data of J. E. Rowe (private communication) for Cl adsorbed on Cd show a resonance ~2 eV wide centered 5.8 eV below E_F, with a shoulder on the low-energy side suggestive of a σ-π splitting of ~0.8 eV. This is consistent with Fig. 3, in conjuction with a bond length deduced from covalent or metallic radii as discussed by P. M. Marcus, J. E. Demuth, and D. W. Jepsen [Surface Sci. 53, 501 (1975)] (which corresponds to a d of ~1 a.u.). The calculated σ component of the resonance for this d is 0.7 eV further below E_F than the π component.

Infrared Cyclotron Resonance in Semiconducting Surface Inversion Layers*

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The frequency-dependent conductivity $\sigma(\omega)$ of a two-dimensional interacting electron gas in the presence of a random array of scattering centers and a dc magnetic field is studied. Electron-electron interaction effects enter $\sigma(\omega)$ through the holomorphic memory function $M(\omega)$ whose imaginary part is the inverse of the frequency-dependent relaxation time $\tau(\omega)$. A qualitative explanation of some of the perplexing results observed in infrared cyclotron resonance, including how electron-electron interactions affect both the fundamental and harmonics, is given.

The concentration-dependent enhancement of the effective mass[1] and g value[2] of electrons in the surface inversion layer of a semiconductor obtained by studying the amplitude of the Shubnikov-de Haas oscillations in the surface conductance has been attributed[3-5] to many-body interactions among the inversion-layer electrons. These observations spurred interest in observing dynamic cyclotron resonance in these systems.[6,7] A simple argument shows that dynamic cyclotron resonance excited by a spatially uniform ac electric field cannot be affected by many-body interactions among the conduction electrons in a free electron gas.[8] For this reason the many-body enhancement observed in the dc experiments would not be expected to occur in infrared cyclotron resonance. Recent experimental observations have yielded some unexpected results.[9-11] The most striking result is the appearance of "harmonics" of cyclotron resonance at not quite integral multiples of the fundamental resonance. The amplitude of harmonics is relatively large (up to the order of 10% of the fundamental in the derivative spectrum) and dependent on the electron concentration. The mass obtained by equating the resonance frequency to an integral multiple of eB/m^*c displays, as a function of concentration, a behavior similar to that of the effective mass observed in the dc experiments. In the absence of electron-electron interactions, impurity scattering leads to the occurrence of "harmonics" at exact multiples of the bare cyclotron frequency.[12] In addi-

tion to the appearance of harmonics, unexplained concentration and frequency dependence of the mass associated with the fundamental resonance has been observed by some authors.[10] These effects have been attributed[10] to electron-electron interactions in the presence of a random array of scattering centers. The fact that the frame of reference of the fixed impurities is a preferred frame of reference renders inapplicable Kohn's argument[8] about the effect of electron-electron interactions.[13] In this paper we present a theory of cyclotron resonance which incorporates these effects. The ac conductivity is found to be $\sigma_\pm = (iNe^2/m)[\omega \mp \omega_c + M(\omega)]^{-1}$, and the function $M(\omega) = M_1 + iM_2$ is evaluated to terms linear in the concentration of impurities. The fundamental resonance occurs at $\omega + M_1(\omega) = \omega_c$, and its width depends on $M_2(\omega)$. The harmonics result from structure in $M(\omega)$ close to multiples of ω_c*, a renormalized cyclotron frequency. In the absence of electron-electron interactions the structure in $M(\omega)$ occurs at multiples of the bare cyclotron frequency ω_c as found by Ando.[12]

In the presence of a dc magnetic field $\vec{B} = (0,0,B)$ oriented normal to the surface, the power transmitted[14] through the surface inversion layer can be expressed in terms of the frequency-dependent conductivity $\sigma_\pm(\omega) = \sigma_{xx}(\omega) \mp i\sigma_{xy}(\omega)$. The conductivity itself can be written as[15]

$$\sigma_{\alpha\beta}(\omega) = \frac{i}{\omega}\frac{Ne^2}{m}\delta_{\alpha\beta} + \frac{i}{\omega}\left(\frac{e}{m}\right)^2 \int_{-\infty}^{\infty} dt\, e^{i\omega t} Q_{\alpha\beta}(t), \tag{1}$$

where $Q_{\alpha\beta}(t)$ is proportional to the current-current correlation function

$$Q_{\alpha\beta}(t) = \langle\langle \Pi_\alpha | \Pi_\beta \rangle\rangle = -i\theta(t)\langle [\Pi_\alpha(t), \Pi_\beta(q)] \rangle. \tag{2}$$

In these equations $\vec{\Pi} = \vec{P} + (Ne/c)\vec{A}$, where M and $\vec{P}$ are the mass and momentum of the center of mass, $\vec{A}(\vec{R})$ is the value of the dc vector potential at the position of the center of mass, and N is the number of electrons in the system (we have taken the surface area to equal unity). The total current operator is simply $\vec{J} = (-Ne/M)\vec{\Pi} = -(e/m)\vec{\Pi}$.

We introduce center-of-mass coordinates $\vec{R} = N^{-1}\sum_j \vec{r}_j$, $\vec{P} = \sum_j \vec{p}_j$ and relative coordinates[16] $\tilde{\vec{r}}_j = \vec{r}_j - \vec{R}$, $\tilde{\vec{p}}_j = \vec{p}_j - N^{-1}\vec{P}$. The Hamiltonian of the system can be separated into three parts:

$$H = H_{\text{c.m.}} + H_R + U. \tag{3}$$

The center-of-mass Hamiltonian is that of a free particle of mass M and charge $-Ne$ in the presence of a uniform magnetic field. H_R is the Hamiltonian for the internal degree of freedom and includes the electron-electron interactions. The electron-impurity Hamiltonian U can be written

$$U = \sum_{q,j,l} u_q \exp\{i\vec{q}\cdot(\vec{R}_l - \tilde{\vec{r}} - \vec{R})\}, \tag{4}$$

where $\vec{R}_l$ is the position of the lth impurity. In the absence of electron-impurity scattering (when $U=0$), the center-of-mass and relative degrees of freedom are completely uncoupled. A spatially uniform ac electric field couples only to the center-of-mass coordinate, so that Kohn's argument[8] is clearly valid. We investigate the effect of impurities by studying the equations of motion of the Green's function $Q_{\alpha\beta}(\omega)$. This yields the following expression for $Q_\pm(\omega)$ (to lowest order in the impurity concentration):

$$Q_\pm(\omega) = \pm\frac{M\omega_c}{\omega + \omega} + \frac{J_{xx}(\omega)}{(\omega \mp \omega_c)^2}, \tag{5}$$

where

$$J_{xx}(\omega) = -4\pi N_I \textstyle\sum_q q_x^2 |u_q|^2 [\chi(q,\omega) - \chi(q,0)] v_q^{-1}, \tag{6}$$

and

$$-4\pi v_q^{-1}\chi(q,\omega) = \langle\langle \rho_q | \rho_{-q} \rangle\rangle_\omega = -i\int dt\, e^{i\omega t}\theta(t)\langle [\rho_q(t), \rho_{-q}(0)] \rangle. \tag{7}$$

In these equations N_I is the impurity concentration $\rho_q = \sum_j \exp(i\vec{q}\cdot\vec{r}_j)$, and v_q is the Fourier transform of the electron-electron interaction.

Equation (6) is valid only to lowest order in N_I. However, the result can be generalized by making

use of the memory function approach of Götze and Wölfe[17] to write the conductivity as

$$\sigma_{\pm}(\omega) = \frac{iNe^2/m}{\omega \mp \omega_c + M(\omega)}. \tag{8}$$

The expression for $M(\omega)$ to lowest order in N_I can be obtained by expanding Eq. (8) to lowest order in $M(\omega)$ and comparing with Eqs. (1), (5), and (6); we find

$$M(\omega) = (mN\omega)^{-1} J_{xx}(\omega). \tag{9}$$

The conductivity can be expressed as

$$\sigma_{\pm}(\omega) = \frac{iNe^2/m^*(\omega)}{[\omega - eB/m^*(\omega)c] + i\tau^{-1}(\omega)}, \tag{10}$$

where $m^*(\omega) = m[1 + \omega^{-1} M_1(\omega)]$ and $\tau(\omega) = M_2^{-1}[1 + \omega^{-1} M_1(\omega)]$. Here M_1 and M_2 are the real and imaginary parts of the memory function $M(\omega)$. Both are proportional to the impurity concentration, and their magnitudes are of the order of the relaxation frequency τ^{-1}. The fundamental resonance occurs at the frequency satisfying the equation $\omega = eB/m^*(\omega)c$, which is renormalized from the bare cyclotron frequency by a factor $[1 + \omega^{-1} M_1(\omega)]^{-1}$. It reduces to ω_c in the limit of zero impurity concentration as required by Kohn's argument.[8]

We demonstrate below how electron-electron interactions influence the harmonic structure. The function $\chi(q, \omega)$ appearing in Eq. (6) has poles at the magnetoplasma modes of the electron gas in the nonretarded limit.[18] For the purpose of illustration we approximate $\chi(q, \omega)$ as a sum over its poles (i.e., a plasmon-pole type approximation). Then

$$-4\pi v_q^{-1} \chi(q, \omega) = \sum_{l=1}^{\infty} \Gamma_l(q) \left[\frac{-1}{\omega + i\delta - \omega_l(q)} + \frac{1}{\omega + i\delta + \omega_l(q)} \right], \tag{11}$$

where $\Gamma_l(q) = -\epsilon_0/4\pi^2 [\partial \chi_0(q, \omega)/\partial \omega]_{\omega = \omega_l(q)}$. Substitution of (11) into (9) and (6) yields

$$M_2(\omega) = \frac{\pi N_I}{mN\omega} \sum_{ql} q_x^2 |u_q|^2 \Gamma_l(q) \delta(\omega - \omega_l(q)). \tag{12}$$

It is clear that $M_2(\omega)$ has a series of maxima near $\omega = \omega_l(q_0)$, where q_0 characterizes the wave vector around which the main contribution to the sum over q in Eq. (12) originates. Using the expression for $\chi_0(q, \omega)$ in Ref. 18, it can easily be shown that $\omega_l(q)$ as a function of $X = qv_f/\omega_c$ is of the form [19]

$$\omega_l^2(q) = \omega_c^2 [l^2 + O(X^{2l-1})] \text{ for } X \ll 1, \tag{13}$$

and

$$\omega_l(q) = l\omega_c + O(X^{-2}) \text{ for } X \gg 1, \tag{14}$$

where l is a positive integer. If one employs the $\chi_0(q, \omega)$ including short-range correlation effects as that obtained in the Fermi-liquid theory,[20] two important changes occur. The first is that the frequency ω_c appearing in Eqs. (13) and (14) is renormalized to $\omega_c^* = (1 + A_1)^{-1} \omega_c$. The second is that the frequency of each mode at $X = 0$ is shifted to $\omega_l = (1 + A_l) l \omega_c^*$. Here A_l is the lth Fourier coefficient of the Landau[21] interaction function.[22] Equations (12)–(14) indicate that $M_2(\omega)$ exhibits peaks near but not exactly at harmonics of ω_c^*. The exact positions of peaks and their linewidths depend on the dispersion relation of the magnetoplasma modes $\omega_l(q)$ and the weighting function $q_x^2 |u(q)|^2 \Gamma_l(q)$. They should vary with the type of impurity and the electron concentration. This structure exists even if we neglect the quantum oscillations.

The model[23] described in this paper thus explains in a qualitative way the surprising experimental results on infrared cyclotron resonance in semiconducting surface inversion layers. We have used the plasmon-pole approximation only for the purpose of illustrating the qualitative features. Detailed calculations of the amplitude and line shape of the resonances for different type impurities, and for a wide range of frequencies, electron concentrations, and impurity concentrations, employing the full $\chi(q, \omega)$, will be presented in a later publication.

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[19]These are solutions of the equation $\epsilon_0+2\pi\,\mathrm{Re}\,\chi_0(q,\omega)=0$. $\chi_0(q,\omega)$ has also an imaginary part consisting of a series of sharp δ-function peaks at multiples of ω_c associated with single-particle excitations. If collision broadening is included in χ, the δ functions are broadened and can lead to additional structure at χ at multiples of ω_c. In a more rigorous formulation of the plasmon-pole approximation, the dispersion of modes should be slightly modified to account for the single-particle excitation.

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