

Effect of Electron-Electron Interactions on Photoemission in Simple Metals*

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Recently interest has arisen in the possibility of significant deviations of the optical properties of metals from their random-phase-approximation (RPA) values. Starting with the approximation that the crystal pseudopotential can be regarded as weak, we have carried out calculations beyond RPA of the optical absorption and primary photoelectron kinetic-energy distribution. Dynamic screening (as opposed to the usual RPA static screening) results in corrections of great importance to the phonon (or disorder) contribution to the optical absorption and makes qualitative changes in the phonon-induced photoelectron distribution for photons having energies above w_p . A new feature is a marked peak in the phonon contribution to the photoelectron distribution at the Fermi energy for photons having energies in the region of plasmon energies. The supposition of a weak crystal potential prevents quantitative application of these results except to Na and K, but the qualitative effect of dynamic screening should be the same in more complicated systems. The possibility of the experimental existence of photoemission features inexplicable in RPA has been recently discussed by Phillips.

I. INTRODUCTION

RECENTLY, attention has been focused on the possibility of the existence of anomalous effects in the optical absorption¹⁻³ and external photoemission⁴ of simple metals. Various lines of theoretical approach³⁻⁷ have been utilized to explain the experimental effects.

Two features are common to these approaches for calculating optical absorption. One feature is that fairly radical departures from the usual Hartree approximation and Hartree dielectric function are employed. The second is that when the optical absorption is calculated in the long-wavelength limit, a sink which takes momentum from the electrons must be found. Overhauser's optical-absorption calculation for the free-electron gas seems to violate the conservation of momentum in its present form as no sink for electron momentum is supplied (see Appendix). Cohen does not carry his calculation of optical absorption out sufficiently far to include such details. His analogies with superconductors would lead one to believe that phonons are the sink for momentum, although the temperature dependence he mentions does not agree with this.

Photoemission studies provide additional information about the nature of optical absorption at a single photon energy through the distribution of photoelectron kinetic energies, and serves, therefore, as a useful tool in understanding the nature of optical absorption. In this paper the effects of calculating the optical absorption for simple metals in the *simplest* approximation

beyond the usual Hartree approximation on the energy distribution of the primary photoelectrons is investigated. It is shown that when the optical absorption is due to electron-phonon scattering or disorder scattering, a sizeable excess of primary photoelectrons originate near the Fermi surface for photon energies near the plasma frequency. This mechanism can provide a simple explanation for photoemission anomalies discussed by Phillips.

In Sec. II, the framework of approximations in which the results are calculated is discussed, and the calculation of the optical absorption is reviewed. The implications of this treatment for photoemission are treated in Sec. III and numerical results presented for potassium. The results are discussed in Sec. IV.

II. THE OPTICAL ABSORPTION

We treat a model of a simple metal in which the electron Fermi sea is regarded as being weakly perturbed by the positive ions in the metal. Each ion is regarded as having associated with it a weak potential $V(r)$. Optical absorption will be calculated only to second order in $V(r)$, the first nonvanishing term.

The actual bare-ion potential is of course not small. In pseudopotential form, there is a near cancellation between large Z effects and inner-shell repulsive orthogonalization terms. Harrison⁸ has noted that for Born-approximation calculations the ion potential can often be represented as

$$V(r) = -[e^2/r - V_0 a^3 \delta^3(r)], \quad (1)$$

where a is the Bohr radius and V_0 is a constant. For backscattering in sodium, Harrison⁹ finds that the Fourier transform of (1) vanishes for a wave vector of $1.6 \times 10^8 \text{ cm}^{-1}$. We shall use this ion potential in numerical calculations. Fortunately, answers are only weakly dependent on V_0 .

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¹ H. Mayer and M. H. El Naby, *Z. Physik* **174**, 289 (1963).

² J. N. Hodgson, *Phys. Letters* **7**, 300 (1963).

³ M. H. Cohen and J. C. Phillips, *Phys. Rev. Letters* **12**, 662 (1964).

⁴ J. C. Phillips, *Phys. Rev.* **137**, A1835 (1965).

⁵ M. H. Cohen, *Phys. Rev. Letters* **12**, 664 (1964).

⁶ A. W. Overhauser, *Phys. Rev. Letters* **13**, 190 (1964).

⁷ A variety of other explanations, including excitons, the anomalous skin effect, and indirect transitions to the zone edges, have also been suggested (chiefly informally) as possible.

⁸ W. A. Harrison, *Phys. Rev.* **136**, A1107 (1964).

⁹ W. A. Harrison (private communication).

The effect of the ion potential on the energy band structure after the potential is screened by the Fermi gas is in fact small in simple monovalent metals like Na and K. This does not guarantee that the potential can be represented by a small pseudopotential for all purposes, but no qualitative errors are presently known from such a representation in these metals.

When electron-phonon coupling is small, the electromagnetic properties at frequencies large compared to phonon frequencies can be calculated as if the ions were fixed instantaneous positions, using the ensemble distribution of positions appropriate to the given system. Such an approximation washes out dynamical electron-phonon coupling effects and zero-point fluctuations, and averages properties over an energy interval $k\Theta_D$, where Θ_D is the Debye temperature. For high-frequency calculations and $T \gg \Theta_D$, the error introduced is negligible.

If one Fourier component of the perturbation potential is written as $V_q \exp i\mathbf{q} \cdot \mathbf{r}$, to calculate optical loss and photoemission we must calculate the contribution to the imaginary part of the dielectric constant of this perturbation. In the absence of all perturbations, denote the eigenstates of a many (interacting) particle electron-gas system by $|\psi_i\rangle$, having energies E_i . These states are also eigenstates of the total momentum, so have quantum numbers P_{xi} also. If an electric field perturbation in the x direction $\mathbf{E} = \mathbf{i}E \cos \omega t$ is included written as a vector potential, the Hamiltonian becomes

$$T_{\text{electrons}} + V_{\text{electron-electron}} + \frac{e}{m\omega} \sum_{\text{electrons } n} (P_n) \cdot \mathbf{i}E \sin \omega t \quad (2)$$

(omitting terms in the square of the vector potential, as we seek a current linear in E). \mathbf{P}_n is the electron momentum operator. The many-particle eigenfunctions $|\psi_i\rangle$ are eigenfunctions of this Hamiltonian, and have the time dependence

$$\exp\{-i[(E_i/\hbar)t - (P_{xi}E/\hbar m\omega^2) \cos \omega t]\}.$$

The current can now be expressed correctly to second order in V_q and first order in E by doing perturbation theory with these basis functions correct to second order in V_q . If, in increasing powers of V_q , the wave function is expanded $\psi = \psi_0 + \psi_1 + \psi_2$, then the current density correct to order V_q^2 is

$$\mathbf{J} = -\frac{e}{m} \langle \psi | \sum_{\text{electron } n} \left(\mathbf{P}_n - \frac{e}{c} \mathbf{A}_n \right) | \psi \rangle = -\left(\frac{N}{\Omega} \right) \frac{e^2}{mc} \langle \psi | \mathbf{A} | \psi \rangle + \frac{e^2}{m} \langle \psi_1 | \mathbf{P} | \psi_1 \rangle, \quad (3)$$

where Ω is the crystal volume.

Since \mathbf{A} contains no spatial variation, its expectation value can be trivially evaluated. Since $|\psi_0\rangle$, the ground

state of the system, is an eigenstate of the momentum having eigenvalue zero, other terms from the left-hand side of (3) vanish in order V_q^2 . By writing out ψ_1 explicitly and summing over the various Fourier components of the potential, one finds (for a system having an isotropic conductivity) that the imaginary part of the dielectric function is

$$\epsilon_2(\omega) = \frac{4\pi^2 e^2}{\Omega m^2 \omega^4} \sum_q \sum_i \left(\frac{P_{xi}}{\hbar} \right)^2 \times |\langle \psi_i | V_q \sum_n e^{i\mathbf{q} \cdot \mathbf{r}_n} | \psi_0 \rangle|^2 \delta(E_i - E_0 - \hbar\omega). \quad (4)$$

Perturbation theory can also be used to express the dielectric shielding of an oscillating nonuniform potential $V(\mathbf{r}, t) = V_0(\cos \mathbf{q} \cdot \mathbf{r}) \cos \omega t$ in the free-electron gas in terms of matrix elements and energies of the exact many-body wave functions. From the definition of the dielectric function in terms of the system response, an expression for the imaginary part of the reciprocal of the longitudinal dielectric function is obtained.

$$\text{Im} \left(\frac{1}{\epsilon(q, \omega)} \right) = \frac{4\pi^2 e^2}{\Omega q^2} \sum_i |\langle \psi_i | \sum_n e^{i\mathbf{q} \cdot \mathbf{r}_n} | \psi_0 \rangle|^2 \times \delta(E_i - E_0 + \hbar\omega). \quad (5)$$

Comparing (4) and (5), one obtains

$$\epsilon_2(\omega) = \frac{1}{m^2 \omega^4} \sum_q |V_q|^2 q_x^2 q^2 [\text{Im}(1/\epsilon(q, \omega))], \quad (6)$$

an expression exact to order V_q^2 including all electron-electron interactions.

This calculation has been made for an electron gas at zero temperature. The smearing of the Fermi surface by thermal excitation is expected to introduce no explicit effects at optical frequencies except perhaps slight broadenings. Equation (6) is not new—it is the expression obtained by Ron and Tzoar¹⁰ for absorption in a quantum plasma. However, the present derivation makes it clear that (6) is exact to order $|V_q|^2$.

A Hartree calculation of $\epsilon_2(\omega)$, as described by Ehrenreich and Cohen,¹¹ would proceed as follows. First, find the self-consistent energy eigenstates of the potential. For a weak potential, the self-consistent potential would be the applied potential with static screening from the electron gas. Second, calculate the response of the system to an applied uniform electric field oscillating in time, keeping self-consistently only the response of the system at wave vector zero. The potential due to the ions is screened statistically.

Let

$$\epsilon^H(\mathbf{q}, \omega) = \epsilon_1^H(\mathbf{q}, \omega) + i\epsilon_2^H(\mathbf{q}, \omega) \quad (7)$$

¹⁰ A. Ron and N. Tzoar, Phys. Rev. **132**, 2800 (1963).

¹¹ H. Ehrenreich and M. H. Cohen, Phys. Rev. **115**, 786 (1959).

be the Hartree longitudinal dielectric function of the free-electron gas. If we approximate $\epsilon(q, \omega)$ in the right side of (6) by $\epsilon^H(q, \omega)$ the quantity in parentheses is

$$(\epsilon_2^H(q, \omega) / |\epsilon_1^H(q, \omega)|^2). \quad (8)$$

This result still describes physical effects not present in simple Hartree calculations.

The Hartree calculation (à la Ehrenreich and Cohen) yields the same answer but with this parenthesis replaced by

$$(\epsilon_2^H(\mathbf{q}, \omega) / |\epsilon_1^H(\mathbf{q}, 0)|^2). \quad (9)$$

For low frequencies ($\omega \ll \omega_p$) or large wave vectors $q \gg (\text{screening length})^{-1}$, the two expressions are the same. For small wave vectors and large frequencies, there is a real physical difference between the Hartree calculation, which includes static screening only, and Eq. (6) [with (8)] which contains dynamic screening effects.

Ron and Tzoar calculated the optical absorption for a plasma from Eq. (6). We can do the same using the atomic potential (1). Define

$$U_q = \int V(r) e^{-i\mathbf{q} \cdot \mathbf{r}} d^3r. \quad (10)$$

Then

$$|V_q|^2_{\text{average}} = |U_q|^2 (N |\Omega^2| \left[1 + \int f(r) e^{-i\mathbf{q} \cdot \mathbf{r}} d^3r \right]), \quad (11)$$

where $f(r)$ is the pair distribution function. The bracket in (11) is, of course, the same factor which appears in x-ray scattering, and can be directly measured.

For a solid, the bracket in (11) consists of two parts. One part, due to lattice periodicity, exists for $\mathbf{q} = 2\pi\mathbf{G}$, where \mathbf{G} is a reciprocal lattice vector. The other part, due to thermal (or structural) disorder is spread approximately uniformly through \mathbf{q} space. For potassium, the absorption shape of the present theory due to the periodic lattice will be very nearly that calculated by Butcher,¹² since even for the smallest reciprocal lattice vector $E_1^H(2\pi\mathbf{G}, \omega)$ is equal to unity within 25%, and $E_2^H(2\pi\mathbf{G}, \omega)$ is small. Experimental data suggests that this term alone cannot explain the bulk of the optical absorption in potassium above 1 eV.

If the bracket in Eq. (11) is represented as a constant for the disorder scattering, the shape of the optical absorption is very like that of Ron and Tzoar. The shape of $\epsilon_2(\omega)$ due to disorder scattering in this approximation is shown in Fig. (1) for potassium. For $T \gg \Theta_D$, the thermal disorder part of the bracket in (11) is proportional to temperature.

III. THE PHOTOELECTRON ENERGY DISTRIBUTION

The primary electrons responsible for photoemission have kinetic energies (above the Fermi energy E_F) between E and $E - E_F$ (or 0 if $E_F > E$), where E is the

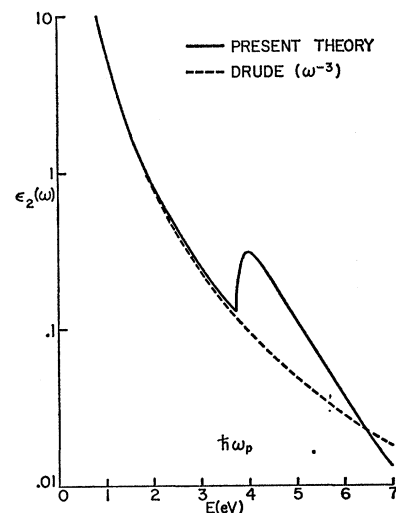


FIG. 1. The phonon disorder part of $\epsilon_2(\omega)$ calculated from Eqs. (6), (8), and (11) as a function of energy for potassium. The ordinate contains an arbitrary multiplicative factor.

photon energy. The problem of connecting the actual photoemission to the kinetic energies of the primary electrons being well-known, only the distribution of kinetic energy of primary electrons will be considered here. Let $E_p(q)$ be the energy of a plasmon of wave vector q . In the Hartree dielectric-function approximation, real plasmons extend from $q=0$ to a critical q_c . The primary electron distribution calculation naturally divides itself into two parts, to be treated separately.

A. $E < E_p(0)$ or $E > E_p(q_c)$

In this energy region, $\text{Im}[\epsilon^H(\mathbf{q}, \omega)]^{-1}$ is due to the excitation of electron-hole pairs. Precisely the same processes occur in the present calculation in this energy interval as in the ordinary Hartree calculation. The relative *weights* assigned to different excitations are, however, greatly changed.

The difference between the present and the ordinary calculation originates from the use of Eq. (8) instead of Eq. (9). For a typical value of q ($q = 1.2k_F$) and parameters appropriate to potassium, Eq. (9) has its largest value at $E = 2.9$ eV. (See Fig. 2.) At this photon energy, the kinetic energy of the electrons generated are distributed uniformly between 0.9 and 2.9 eV above the Fermi energy. (The Fermi energy of potassium is 2.05 eV.) Equation (8) on the other hand has its maximum at $E = 7.0$ eV. At this photon energy, the kinetic energy of the electrons generated are distributed between 6.0 and 7.0 eV. Said slightly differently, Eq. (9) for this q excites electrons from states within ~ 2 eV of the Fermi energy, while Eq. (8) excites them from states within only ~ 1 eV of the Fermi energy.

Detailed calculations are given in Fig. 3. It is most convenient to plot the energy of the state of *origin* of the final electron, whose final total energy is this plus the photon energy. At low photon energies, the curve approaches that to be expected for the ordinary Hartree calculation, namely the parabolic free-electron density

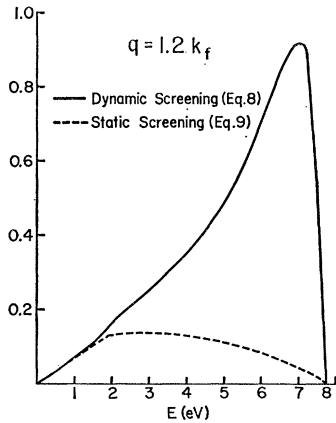


FIG. 2. A comparison between expression (8) (dynamic screening) and expression (9) (static screening) for $q = 1.2k_f$ in potassium.

of states. At high photon energies, a definite emphasis on states near the Fermi surface is clearly observable, to be expected from the qualitative considerations of the previous paragraph.

B. $E_p(0) < E < E_p(q_c)$

In this energy region, $\text{Im}[\epsilon^H(q, \omega)]^{-1}$ is due to two separate processes. One of these is the generation of electron-hole pairs as in A, and can be treated in the same fashion. The other process is the generation of real plasmons. The problem which remains is to calculate the primary electrons which optical-absorption generating real (Hartree) plasmons will introduce.

A plasmon generated in the initial absorption process will break up into quasiparticle excitations by two mechanisms. First, if a more exact calculation of properties of the electron gas is made, the real (Hartree) plasmon is unstable against decay into four quasiparticles. The lifetime for such decay is a function of the plasmon wave vector, becoming infinite in the limit $q \rightarrow 0$. Such processes will produce electrons having an energy distribution like those which have suffered a major inelastic collision, as the total photon energy is partitioned between two pairs of quasiparticles instead of one pair.

Second, if the crystal potential were included in higher order, the plasmon could scatter against the crystal potential and decay into an electron-hole pair. This process takes place even for $q=0$ plasmons. Thus, for any given size of crystal potential, eventually for small enough q the crystal potential causes the plasmon to decay.

The breakup of the plasmon by the lattice potential is easily calculated in lowest order. The (Hartree) plasmon creation operator can be taken as

$$A \sum \sum \frac{c_{k+q}^\dagger c_k}{E_p(q) + E_k - E_{k+q}}, \quad (12)$$

$$|\mathbf{k}| < k_F,$$

$$|\mathbf{k} + \mathbf{q}| > k_F.$$

If the lattice introduces some effective one-body potential $V(r)$, it can, in lowest order, scatter either the electron or the hole, but not both. For plasmon wave vectors such that

$$\hbar^2/2m(k_F + q)^2 < E_p(q) \quad (13)$$

hole scattering cannot conserve energy, so only electron scattering is available. Electron scattering from pair component of the plasmon takes place independently. Restricting ourselves to q which satisfy (13), ($q < 0.35k_F$ for potassium), the probability of leaving a hole in state k is then proportional to

$$[1/(E_p(q) + E_k - E_{k+q})]^2 W(E_{k+q} \rightarrow E_p(q) - E_k). \quad (14)$$

Here $W(E_{k+q} \rightarrow E_p(q) - E_k)$ is the probability per unit time that an electron placed in state $\mathbf{k} + \mathbf{q}$ but arbitrarily assigned an energy $E_p(q) - E_k$ would be scattered by the lattice into a final state of energy $E_p(q) - E_k$. W , of course, depends on the lattice potential, but has a form which depends only weakly on the form of the lattice potential. For a potential with uniform Fourier components (much like the screened potential of the present problem) W will vary as $[E_p(q) - E_k]^{1/2}$, a dependence weak enough to be neglected in the present calculation.

Putting $W = \text{constant}$ in (14), the probability of leaving a hole in state k can be calculated from (12) by calculating the normalizing factor in the probability distribution. The electron then has the total energy $E_k + E_p(q)$.

The plasmons generated by photons of energy E must have the same energy: i.e., $E_p(q) = E$. By choosing E only slightly above $E_p(0)$, q can be kept small, making the lattice potential the dominant source of plasmon decay into particles. For such an E , Eq. (13) would also be satisfied. [In potassium (13) is satisfied from $E_p(0) = 3.7$ to 3.9 eV.]

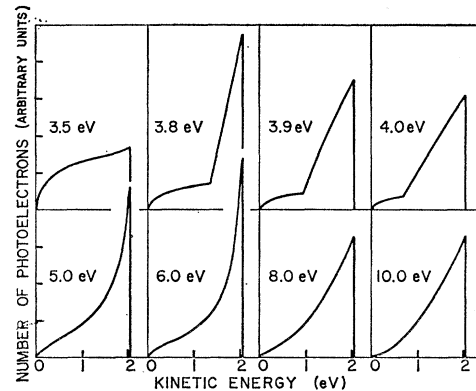


FIG. 3. The primary photoelectron kinetic-energy distribution from that part of the optical absorption due to phonons (or disorder). Each curve is normalized to the same area. The energy zero is chosen as $E_{\text{photon}} - E_{\text{Fermi}}$, the minimum kinetic energy possible above the Fermi energy for a given photon energy. In the Hartree approximation, all curves would resemble a free-electron parabola. The photon energies in electron volts are given in the upper left corners.

It is not inconsistent with the original spirit of the calculation to order $|V_q|^2$ to include damping due to the lattice potential. This damping introduces no added absorptions: It only ensures that the absorption states are allowed to decay to the final products important in photoemission. The entire photoemission electron distribution is still proportional only to $|V_q|^2$. Calculated photoemission electron distributions for this case are also given in Fig. 3.

For photon energies and disorder wave vectors above the hole-scattering threshold, a small amount of hole scattering will occur. Even for the worst energy, $E \approx E_p(q_c)$, the low density and consequent electron-hole asymmetry make the electron scattering about three times more important than the hole scattering. (The real plasmon contribution goes smoothly to zero at this energy.) Equation (14) is then a fair approximation at all photon energies.

IV. DISCUSSION

The optical absorption of potassium is as yet poorly understood. When this very simple metal is not well understood, basic questions about the interpretations given in more complicated systems arise. Photoemission studies provide a good complement to optical-absorption studies, for even at a single photon energy information can be obtained about the nature of the electronic transitions. The calculation presented shows that the primary photoelectron energy spectrum in simple metals shows significant additional structure when higher approximations than Hartree are used to calculate the optical absorption, and absorption is due to disorder.

Phillips⁴ has recently pointed out that there is some experimental evidence in potassium for an unexpectedly large number of photoelectrons originating near the Fermi surface for some energies of excitation. He has interpreted this in terms of an unspecified form of "collective resonance." An alternative explanation of such a result is that the excess is just what would be expected from the non-Hartree calculation of photoelectrons due to disorder, since the critical range of photon energies seems to be at and above the plasma energy as in Fig. 3.

The present theory is clearly not quantitatively applicable to Ag or Cu. A similar qualitative effect would be expected, however, especially in silver which has a strong plasmon¹³ at 3.8 eV for $k=0$. Phillips notes that a large excess of photoelectrons from the Fermi surface occurs for photons of energies near 3.8 eV. Owing to band-structure differences copper does not (at least at $k=0$) have a recognizable plasmon.¹³ It does not display the strong photoemission effect shown in silver.

It is somewhat surprising to note in Fig. 3 that the

free-electron parabola, well approximated at low photon energies, is not returned to by 10.0 eV. The shape at this photon energy is still dominated by small wave-vector components of the potential, which are well screened at low frequencies but unscreened at high frequencies.

The calculated qualitative effect of corrections to the Hartree-approximation optical properties on photoemission ought to exist (at least in simple metals like potassium) if the ordinary free-electron ground state is qualitatively correct. In potassium and perhaps sodium the possibility of having the disorder contribution completely swamped by the lattice contribution is minimized. The careful study of the temperature dependence of the photoelectron energy distribution for photon energies near and above the plasmon energy would be most useful in separating disorder and periodic contributions to look for the predicted effect.

If the ground state in potassium is free electron like, the most major error in the present calculation is the supposition that the crystal potential is small. Indeed, the crystal potential is not small—only its effect on the free-electron energy levels near the Fermi energy is small. Errors due to the magnitude of the crystal potential might introduce additional structure, but there is no reason to expect them to eliminate the qualitative effect calculated here. The use of the Hartree dielectron function limits the quantitative reliability of these calculations, but should not be in qualitative error. Calculations of the optical properties of potassium, if the crystal potential is not regarded as small, are now being attempted.

V. ACKNOWLEDGMENT

The author is indebted to R. A. Ferrell for stimulating his interest in the puzzle of the alkali metals. He would also like to acknowledge a helpful discussion of Ref. 4 with J. C. Phillips prior to its publication.

APPENDIX

Overhauser^{6,14} has calculated the spin-density wave properties of the free-electron gas containing electron-electron interactions but no lattice. In a free-electron gas, electron-electron collisions conserve momentum (and thus current), and therefore cannot result in a real part of the electrical conductivity for uniform fields.

In Overhauser's calculation, there is a periodic potential which is effectively used as a lattice potential for inducing electron band-to-band transitions. When the periodic potential is due to the lattice, this calculation is correct. When this potential is due to the electrons, however, the effect of this potential is also electric-field dependent, and the "backflow" must be calculated. For a uniform electric field, it will precisely cancel the term calculated by Overhauser.

¹² P. N. Butcher, Proc. Phys. Soc. (London) A64, 765 (1951).

¹³ H. Ehrenreich and H. R. Phillip, Phys. Rev. 128, 1622 (1962).

¹⁴ A. W. Overhauser, Phys. Rev. 128, 1437 (1962).

An analogous cancellation is well-known in free-electron spin resonance of alkali metals. In the presence of a magnetic field, the electron gas is spin polarized. An exchange field thus exists which alters the energy required to flip the spin of a particular electron. This exchange field is analogous to Overhauser's periodic potential. If the spin-resonance frequency is calculated without including back flow, it is altered by the exchange field. In actual fact, since the total spin angular momentum commutes with the Hamiltonian, exchange does not alter the spin-resonance frequency, and "backflow" precisely cancels the exchange correction to the spin-resonance frequency. In both cases the cancellation is a necessary consequence of the observables in question, the current and spin, respectively, commuting with the total Hamiltonian.

Under some circumstances, "at $k=0$ " and "the limit

$k \rightarrow 0$ " are not the same in the presence of long-range interactions. In both Overhauser's case and the case of the spin in an electron gas, the relevant excitation has no charge-density fluctuation in the limit $k \rightarrow 0$, so "the limit $k \rightarrow 0$ " and "at $k=0$ " are synonymous. [Overhauser uses a $k \rightarrow 0$ limiting process, but only to show the relation between the power absorption and $\epsilon_2(\omega)$].

This discussion does not bear on the correctness of the spin-density wave ground state in potassium. Were potassium to have such a ground state, other mechanisms (pinning of the spin-density wave or anomalous skin effect) could result in optical absorption displaying the spin-density wave. The Overhauser calculation, however, of a one-parameter optical-absorption shape based on a free-electron gas and the long-wavelength limit seems to be incorrect.

Nuclear Magnetic Resonance of ^{61}Ni in Nickel Metal*

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The zero-applied-field nuclear magnetic resonance of ^{61}Ni in high purity, well-annealed Ni metal has been investigated at room temperature. The technique of rotary saturation is used to obtain a value for the domain-wall enhancement factor $\eta=1600$. Fast passage and saturation effects are observed and interpreted according to theories developed by Portis for inhomogeneously broadened spin systems. This analysis leads to a value for the longitudinal relaxation time $T_1=0.16$ msec, and to an observed nuclear dispersion-to-absorption ratio in the power absorbed of $\beta_0=0.5$ at low rf levels. Fast-passage effects are used to obtain a tracing of the distribution of nuclear magnetic fields in the sample.

INTRODUCTION

ZERO-applied-field nuclear magnetic resonance signals observed at low rf levels in the ferromagnetic metals, Co,^{1,2} Ni,^{3,4} and Fe,⁵⁻⁸ arise from nuclei in the domain walls of the sample. Because of the motion of the domain walls under the influence of the applied rf magnetic field, the rf-field amplitude at the site of

a nucleus in the wall is enhanced by a factor $\sim 10^3$ over the applied field. The strength of the observed signal is also enhanced. Because of a modulation of the intrinsic losses in the sample by the nuclear susceptibility, the detection of the resonance with spectrometers sensitive only to power absorbed leads to signals which are mixtures of nuclear absorption and dispersion. Spin echo experiments and other less direct techniques have shown that these lines are inhomogeneously broadened.^{9,10} Rapid-passage effects have been observed and used to interpret some of the characteristics of the resonances in Co¹ and Fe.⁸

This paper presents the results of an investigation of the nuclear magnetic resonance of ^{61}Ni in pure, well-annealed, unenriched Ni powder at room temperature. The technique of rotary saturation¹¹ is used to find directly a value for the domain-wall enhancement

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