Theory of the perpendicular-field cyclotron-resonance anomaly in potassium

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A simple metal, having a spherical Fermi surface, should not exhibit cyclotron resonance when the magnetic field is perpendicular to the surface. Nevertheless, a sharp resonance was observed by Grimes in potassium. This phenomenon can be explained by a charge-density-wave (CDW) broken symmetry. A small cylindrical piece of Fermi surface, bounded by the CDW gap and the first minigap, contains electrons having very small velocity. These electrons provide a mechanism for the anomalous resonance even though their relative concentration is only $\sim 4\times 10^{-4}$. This same group of electrons is responsible for the sharp photoemission peak (reported by Jensen and Plummer) from (110) surfaces of Na and K.

I. INTRODUCTION

Cyclotron resonance in metals is ordinarily studied in the Azbel-Kaner configuration,[1] for which the dc magnetic field is parallel to the surface. In addition to the fundamental resonance (at H_c), there appears a sequence of subharmonic resonances at H_c/n, $n=2,3,\ldots$. This phenomenon was observed in the simple metals Na and K by Grimes and Kip.[2] Theoretical studies[3] of the surface impedance of an ideal metal, having a conduction-electron energy spectrum $E(\mathbf{k})$ that is parabolic and spherically symmetric, account for the main features of the data and allow accurate determination of the cyclotron mass ($m^*/m=1.24$ and 1.21 for Na and K).[2]

Cyclotron resonance is *not* expected when the magnetic field is oriented perpendicular to the surface.[4] The theoretical curve of Fig. 1 illustrates the variation of the surface impedance with magnetic field for a circularly polarized microwave signal. (The sign convention adopted here corresponds to resonance when $\omega_c/\omega=-1$; $\omega_c\equiv eH/m^*c$.) Structure at resonance should not appear because electrons traveling at the Fermi velocity ($v_F\sim 10^8$ cm/sec) spend too short a time in the skin depth ($\delta\sim 10^{-5}$ cm) compared to the period $2\pi/\omega$ of the microwave field. Electrons near the metal surface travel in and out along (dc) magnetic field lines perpendicular to the surface.

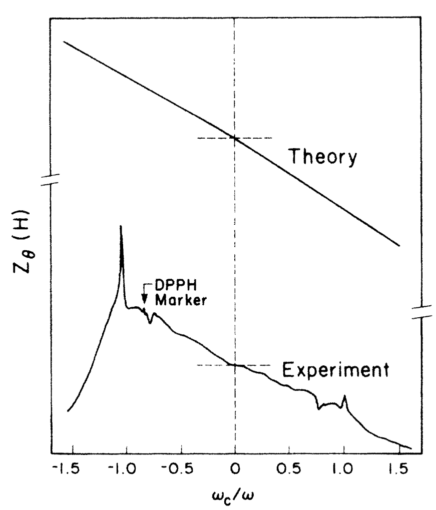

FIG. 1. Surface impedance of potassium vs magnetic field for circularly polarized radiation at $T=2.5$ K. $\omega/2\pi=23.9$ GHz. The theoretical curve is for an ideal metal (spherical Fermi surface) with a specular surface. $\omega_c\equiv eH/m^*c$.

Nevertheless, Grimes found[5] a very sharp *and large* resonance in potassium. This is shown in Fig. 1. The amplitude of the sharp feature is comparable to the total change in surface impedance, $Z(H)$, between $H=0$ and $H=H_c$. From our analysis in Sec. IV the sharpness corresponds to $\omega_\tau\sim 60$, where τ is the electron relaxation time. The DPPH (diphenylpicrylhydrazyl free radical) marker indicates where cyclotron resonance could occur if m^*/m were unity.

The sharp feature shown in Fig. 1 has been attributed[6] to excitation of a Fermi-liquid mode[7] described by an ellipsoidal deformation of the Fermi surface. However, this interpretation has a serious difficulty. The $l=2$ Fermi-liquid mode does not couple to an electric field (which stimulates only $l=1$ Fermi-surface distortions). The $l=2$ mode can arise only from a steep spatial gradient in the electron distribution function $f(\mathbf{k},\mathbf{r},t)$. Variations which occur in the skin depth are too small by 4 orders of magnitude to explain the observed resonance.[6] When one assumes diffuse reflection of electrons at the surface (instead of specular) the shortfall is alleviated by about 1 order of magnitude.[8]

The existence of this resonance anomaly remains enigmatic. Effects of surface roughness (on a scale larger than that required for diffuse scattering) were investigated.[9] The model postulated was represented by a very short relaxation time, $\omega\tau\sim 0.1$, within a distance $\sim\frac{1}{3}\delta$ from the surface. At greater depths a bulk value $\omega\tau\sim 10$ was considered typical. The sharp spatial discontinuity of $\omega\tau$ did lead to a significantly larger coupling to the $l=2$ Fermi-liquid mode. However, there appears to be experimental evidence for discounting this possible explanation.

The samples employed by Grimes were similar[10] to those used in (parallel-field) cyclotron-resonance studies.[2] Well-developed subharmonic resonances were observed, and these required $\omega\tau\sim 10$. Grimes has kindly sent us data which show that the sharp resonance (in perpendicular field) and the Azbel-Kaner signal (in parallel field) occur in *the same specimen.* Subharmonics out to $n=8$

were visible. Comparison with the theoretical expression[11] for the Azbel-Kaner signal indicates that $\omega\tau$ was between 10 and 15. In fact, the $n=2$ subharmonic would disappear if $\omega\tau<3$, and the fundamental would disappear at $\omega\tau=1$.[12] We believe, therefore, that high-order Azbel-Kaner subharmonics cannot be expected in samples when $\omega\tau \ll 1$ in a significant fraction of the skin depth. We therefore propose an alternative theory of the perpendicular-field anomaly, an idea that acquires plausibility from recent photoemission studies of Na and K.[13]

II. CHARGE-DENSITY-WAVE STRUCTURE AND THE FERMI SURFACE

The evidence for a broken translational symmetry of the charge-density-wave (CDW) type in Na and K is extensive.[14] One of the most striking observations is the large number of open-orbit magnetoresistance peaks found in potassium.[15] The origin of this phenomenon is the existence of several small energy gaps in $E(\mathbf{k})$, which cut through the Fermi surface, as shown in Fig. 2. The minigaps and heterodyne gaps arise naturally in the solution of a Schrödinger equation having two incommensurate periodic potentials.[16] Figure 2 is schematic; the separations between adjacent high-order gaps have been exaggerated, and the Fermi-surface distortion is not shown. The heterodyne gaps are very small, $\sim 1-10$ meV, and can suffer magnetic breakdown. The theory of the open-orbit magnetoresistance spectrum[17] provides a rather complete account of the observations.[15] It is worth noting that without a CDW structure the Fermi surface would be simply connected and open orbits could not even occur.

The exact shape of the Fermi surface near the main CDW energy gap cannot be predicted because nonlocal many-body corrections[18] to $E(\mathbf{k})$ are important and are difficult to calculate, especially for a nonuniform electronic charge density. That the Fermi surface comes close to the CDW energy gap is known from the shape (near threshold) of the Mayer—El Naby optical absorption.[19]

There is now direct evidence that a small cylindrical Fermi-surface sheet exists between the first minigap (~ 0.1 eV) and the main CDW gap (0.6 eV), as shown in Fig. 3. Solution[20] of the Schrödinger equation for $E(k_z)$ in the region *between* these two gaps leads to an $E(k_z)$ that is almost a constant, E_c. The relevant question is whether E_c is above or below E_F, the Fermi energy. If $E_c<E_F$, there will be a small cylindrical volume of occupied states (between the two gaps) and these will give rise to a sharp photo-emission peak (normal to a [110] metal surface) at photon energies for which (otherwise) photoemission is forbidden.[20] Such a photoemission signal has recently been reported for Na and K.[13]

Electrons on the cylindrical Fermi surface will have $v_z \ll v_F$, and will accordingly remain in the skin depth for a much longer time than other Fermi-surface electrons. We should remark here that the $\hat{z}$ direction (the direction of the CDW wave vector $\mathbf{Q}$) is known from optical studies to be perpendicular to the surface of a thin film[21] or of a thin slab grown between smooth, amorphous surfaces (from studies of Doppler-shifted cyclotron resonance[22]).

The purpose of the present work is to show that this "new" group of electrons, even though their number is extremely small, can account for the existence and size of the perpendicular-field cyclotron-resonance anomaly, shown in Fig. 1.

In Sec. III we shall calculate the surface impedance $Z(H)$ for a metal having both a standard spherical Fermi surface *and* a small cylindrical sheet (with its axis parallel to the magnetic field $\mathbf{H}$). It is obvious from Fig. 2 that the theoretical model used in what follows greatly oversimplifies the full reality of the CDW structure. Not only do we ignore effects of other high-order gaps, but we neglect the small angle between $\mathbf{Q}$ and the [110] reciprocal-lattice vector.[23] This deviation will cause the

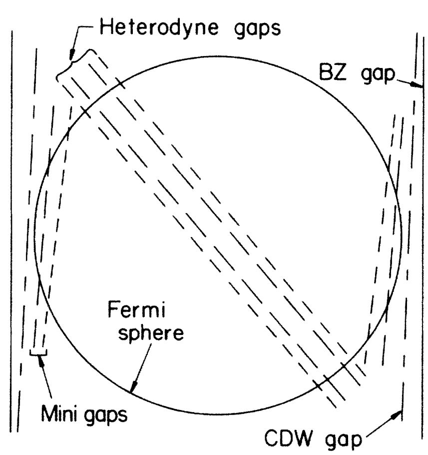

FIG. 2. Schematic illustration of some of the energy gaps that arise (in an alkali metal) from a CDW broken symmetry. The undistorted Fermi sphere is shown. (Spacings between adjacent high-order gaps have been exaggerated.)

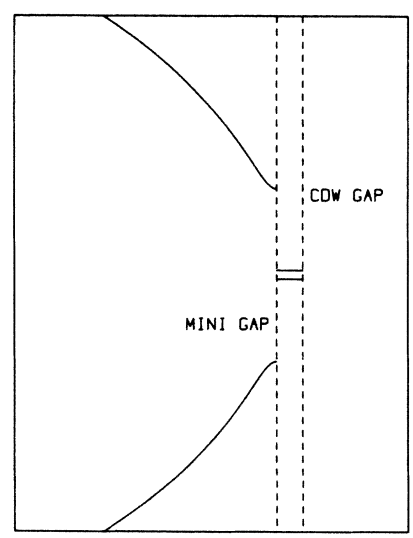

FIG. 3. Sketch of the Fermi surface near the CDW energy gap of potassium. For simplicity the CDW $\mathbf{Q}$ and the reciprocal-lattice vector $\mathbf{G}_{110}$ have been taken parallel, although a small tilt ($\sim 1°$) is anticipated. The small cylindrical sheet of Fermi surface between the CDW gap and the (first) minigap is the focus of this study. (The horizontal axis is parallel to $\hat{\mathbf{z}}$.)

cylinder axis to be tilted away from $\hat{z}$. However, the cyclotron frequency on the cylinder (and for motion perpendicular to $\hat{z}$) is probably not seriously affected. Finally, we remark that the cylinder radius will be taken constant (independent of k_z), which corresponds to $E(k_z)=E_c$. It is clear that the cylinder's radius will actually be slightly larger at the minigap plane compared to its radius at the CDW gap plane. Neglect of this variation is equivalent to taking $v_z=0$ for electrons in the cylinder. Therefore, if such electrons are in the rf skin depth, they remain there during their relaxation time. It follows that a very small concentration suffices to explain the large, anomalous resonance. There are, of course, two cylinders: one near $k_z=\frac{1}{2}Q$ and another near $-\frac{1}{2}Q$.

III. THEORY OF THE SURFACE IMPEDANCE

In accordance with the foregoing discussion, we shall calculate the surface impedance $Z(H)$ for a metal having both a spherical Fermi surface and a small cylindrical one. The axis of the latter will be parallel to $\mathbf{H}$ and perpendicular to the surface. The infinite-medium conductivity $\sigma(q,\omega)$ can be written as the sum of two contributions, one from each group,

$$\sigma=\sigma_s+\sigma_c \ . \tag{1}$$

We shall follow standard procedures[24] to evaluate the conductivity σ_s of the sphere by solving the Boltzmann equation for $f(\mathbf{k},\mathbf{r},t)$,

$$\frac{\partial f}{\partial t}+\mathbf{v}\cdot\nabla_{\mathbf{r}}f-\frac{e}{\hbar}\left[\mathbf{E}+\frac{\mathbf{v}}{c}\times\mathbf{H}\right]\cdot\nabla_{\mathbf{k}}f=\frac{f_0-f}{\tau} \ , \tag{2}$$

where $f_0(\mathbf{k})$ is the equilibrium Fermi-Dirac distribution. We are interested in deviations from f_0 to first order in the electric field,

$$\mathbf{E}\exp(i\mathbf{q}\cdot\mathbf{r}-i\omega t) \ . \tag{3}$$

Accordingly, one looks for a solution having the form

$$f=f_0+f_1\exp(i\mathbf{q}\cdot\mathbf{r}-i\omega t) \ . \tag{4}$$

It follows from Eqs. (2)–(4) that f_1 must satisfy

$$\left[\frac{1}{\tau}+i(\mathbf{q}\cdot\mathbf{v}-\omega)\right]f_1+\frac{eH}{\hbar c}(\mathbf{v}\times\nabla_{\mathbf{k}})f_1=e\mathbf{E}\cdot\mathbf{v}\frac{\partial f_0}{\partial \varepsilon} \ , \tag{5}$$

where $\varepsilon=\hbar^2k^2/2m^*$. It is convenient to change coordinates from $\mathbf{k}$ to ε,k_z,ϕ, where

$$\phi\equiv\tan^{-1}(k_y/k_x) \ . \tag{6}$$

ϕ is the azimuthal angle of an electron in its (k-space) cyclotron orbit. Equation (5) then becomes

$$\left[\frac{1}{\tau}+i(\mathbf{q}\cdot\mathbf{v}-\omega)\right]f_1+\omega_c\frac{\partial f_1}{\partial\phi}=e\mathbf{E}\cdot\mathbf{v}\frac{\partial f_0}{\partial\varepsilon} \ . \tag{7}$$

Equation (7) is easily solved when the propagation vector $\mathbf{q}$ is parallel to $\hat{z}$, which is the case of interest here.

$$f_1=\frac{e}{\omega_c}\frac{\partial f_0}{\partial\varepsilon}\int_{-\infty}^{\phi}d\phi'\,\mathbf{E}\cdot\mathbf{v}\times\exp\left[\frac{[1+i\tau(qv_z-\omega)]}{\omega_c\tau}(\phi'-\phi)\right] \ . \tag{8}$$

The current density $\mathbf{j}(\mathbf{r},t)$ can now be evaluated:

$$\mathbf{j}=-\frac{2e}{8\pi^3}\int f_1\mathbf{v}\,d^3k \ . \tag{9}$$

In tensor notation,

$$j_i=\sum_m\sigma_{im}E_m \ . \tag{10}$$

The tensor components of conductivity (from the sphere) are

$$\sigma_{im}=-\frac{2e\hbar m^*}{\omega_c(2\pi)^3}\int d\varepsilon\frac{\partial f_0}{\partial\varepsilon}\int dk_z\int_0^{2\pi}d\phi\,v_i(\varepsilon,k_z,\phi)\int_{-\infty}^{\phi}d\phi'\,v_m(\varepsilon,k_z,\phi)\exp\left[\frac{[1+i\tau(qv_z-\omega)]}{\omega_c\tau}(\phi'-\phi)\right] \ . \tag{11}$$

For circular polarization of $\mathbf{E}$ we define

$$\sigma_s^+\equiv\sigma_{xx}-i\sigma_{xy} \ . \tag{12}$$

Evaluation of Eq. (11) then leads to

$$\sigma_s^+=\frac{3ne^2\tau}{4m^*}\int_0^{\pi}d\theta\frac{\sin^3\theta}{1-i(\omega+\omega_c)\tau+iql\cos\theta} \ , \tag{13}$$

where n is the electron density and l is the mean free path, $v_F\tau$. ($\cos\theta=k_z/k_F$.) The integration in Eq. (13) is elementary and yields

$$\sigma_s^+(q,\omega)=\frac{3ne^2\tau}{2m^*x^2}\{2ap-1+r(x^2+1-a^2)+i[a+p(x^2+1-a^2)-2ar]\} \ , \tag{14}$$

where

$$a\equiv(\omega+\omega_c)\tau \ ,$$

$$x\equiv ql \ ,$$

$$p\equiv\frac{1}{4x}\ln\left[\frac{1+(x+a)^2}{1+(x-a)^2}\right] \ ,$$

$$r\equiv\frac{1}{2x}[\tan^{-1}(x+a)+\tan^{-1}(x-a)] \ .$$

We next consider the conductivity from electrons in the small cylindrical section of the Fermi sea, shown in Fig. 3. We shall assume that the total number of "cylindrical" electrons is ηn. For the reasons given at the end of Sec. II, v_z for these electrons is neglected. It follows that the transverse conductivity is then the same as in a local theory:

$$\sigma_c^+(\omega)=\frac{\eta n e^2\tau}{m^*}\left[\frac{1}{1-i\tau(\omega+\omega_c)}\right] . \quad (15)$$

(We have assumed that the effective mass m^* is the same for the x,y components of motion on the cylinder as it is for the sphere.)

The surface impedance Z, which is the ratio of the tangential electric field at the surface to the total current density inside the metal, characterizes the microwave-metal interaction.[25] For specular reflection (of the electrons) at the surface, Z^+ is related to the total conductivity,[26] the sum of (14) and (15),

$$\sigma^+(q,\omega)=\sigma_s^+(q,\omega)+\sigma_c^+(\omega) , \quad (16)$$

by

$$Z^+=-8i\omega\int_0^\infty \frac{dq}{c^2q^2-4\pi i\omega\sigma^+} \quad (17)$$

(where c is the velocity of light). The integration in Eq. (17) can only be done numerically.

There are essentially only two unknown parameters in σ^+:$\omega\tau$ and η. We shall adjust $\omega\tau$ so that the width of the sharp resonance is reproduced. The fraction of electrons in the cylinder determines the (vertical) size of the resonance, compared to the change in Z between $H=0$ and H_c. The values we find are $\omega\tau\sim 60$ and $\eta\sim 4\times 10^{-4}$.

IV. RESULTS AND DISCUSSION

Before fitting the theory (just derived) to the data of Fig. 1 it is necessary to discuss in detail several experimental details. The microwave bridge used by Grimes was one built a number of years before by Galt *et al.*[27] It did not incorporate feedback tuning of the klystron to the center of the microwave-cavity resonance. As a consequence, the bridge response corresponded to an unknown linear combination of surface resistance and reactance, which we shall call Z_θ:

$$Z_\theta \equiv \cos\theta\,\mathrm{Re}(Z^+)+\sin\theta\,\mathrm{Im}(Z^+) . \quad (18)$$

The early work on Bi produced such robust signals that the microwave cavity was rhodium plated to reduce the cavity Q.[27] This precaution tends to reduce any effect of detuning, so that the bridge output would then be a measure of surface resistance.

Grimes plated the cavity with high-purity copper to maximize Q and optimize sensitivity.[28] Effects of thermal expansion, magnetoresistance, etc. can result in an unknown (and nonconstant) value of θ during, for example, sweeping of the magnetic field. Another effect of the plating can be seen in the data of Fig. 1. The broad resonant dips near $\omega_c/\omega\sim\pm 0.8$ were probably caused[28] by hydrated $CuSO_4$ particles trapped in the copper plating.[29] (Their resonance frequency corresponds to $g\sim 2.15$.) Finally, the relative sizes of the sharp resonances near $\omega_c/\omega\sim\pm 1$ indicate that the circular polarization was $\sim 90\%$. The bridge was balanced at $H=0$, so the signal shown in Fig. 1 is the change in Z_θ with H, relative to its *unknown* value at $H=0$. The vertical scale factor is also unknown.

Theoretical results from Eqs. (16) and (17) are shown in Fig. 4 for three values of θ. The sharp feature (at cyclotron resonance) is reproduced rather well with $\theta\sim 47°$, $\omega\tau\sim 60$, and $\eta\sim 4\times 10^{-4}$.

One aspect of the data still unexplained is the smooth falloff of Z_θ for magnetic fields beyond resonance. It is, of course, possible that other types of orbits, created by the heterodyne gaps shown in Fig. 2, may be responsible. We have found no way to incorporate such orbits quantitatively.

It is of interest to estimate the radius R of the cylinder having a volume equivalent to the value of η given above. The result depends on the magnitude of the CDW wave vector Q, which is now precisely known. We shall take (from unpublished diffraction data)[30]

$$Q=0.985G_{110} , \quad (19)$$

where G_{110} is the (smallest) reciprocal-lattice vector. It follows that the *total* length L of the two cylinders is[31]

$$L=G_{110}-Q=0.015G_{110} . \quad (20)$$

We have, of course,

$$\pi R^2 L=\frac{4\pi}{3}\eta k_F^3 . \quad (21)$$

From Eqs. (20) and (21) and the fact that $G_{110}=2.3k_F$,

$$R\sim 0.1k_F . \quad (22)$$

Thus the cross-sectional area of the cylinder (perpendicular to $\hat{z}$) is about 100 times smaller than the cross section of the Fermi sphere.

It is of interest to note that Baraff[9] surmised that the sharp cyclotron resonance found by Grimes could be explained by $\sim\frac{1}{20}$ of a monolayer of electrons confined to the surface (and free to move parallel to the surface). He

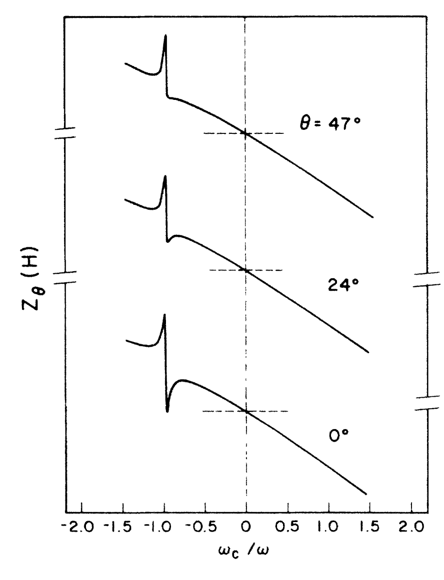

FIG. 4. Theoretical surface impedance $Z_\theta(H)$ for potassium with a CDW broken symmetry. The mixing angle θ (for surface resistance and reactance) is defined by Eq. (18).

emphasized that it was highly unlikely that such a *specularly* reflecting thin film could be formed on the surface of the sample; only it was interesting that so few electrons were needed to give the observed magnitude of the peak. It is not a coincidence that the number of CDW cylinder electrons within a skin depth δ of the surface would constitute $\sim\frac{1}{20}$ of a monolayer equivalent. In other words, the CDW structure provides a small group of electrons, having local properties, that fulfill the essential requirements of Baraff's imagined specular film.

V. CONCLUSIONS

A cylindrical sheet of electrons caused by the CDW broken symmetry of potassium explains the sharp perpendicular-field cyclotron-resonance anomaly found by Grimes. This same group of conduction electrons is responsible[20] for the angle-resolved photoemission anomaly observed by Jensen and Plummer.[13]

The foregoing conclusion can be tested by searching for de Haas—van Alphen or Shubnikov—de Haas oscillations having an oscillation period about 100 times larger than the traditional one. Samples with an oriented CDW wave vector are needed and can probably be obtained with evaporated films. In this case, resistance oscillations (versus H) would likely be the easiest method.

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1M. Ya. Azbel and E. A. Kaner, Zh. Eksp. Teor. Fiz. **32**, 896 (1957) [Sov. Phys.—JETP **5**, 730 (1957)].

2C. C. Grimes and A. F. Kip, Phys. Rev. **132**, 1991 (1963).

3D. C. Mattis and G. Dresselhaus, Phys. Rev. **111**, 403 (1958).

4R. G. Chambers, Philos. Mag. **1**, 459 (1965).

5C. C. Grimes (private communication). The data shown in Fig. 1 were taken in 1966. Similar data (due to Grimes) were published in Ref. 6 and 9.

6G. A. Baraff, C. C. Grimes, and P. M. Platzman, Phys. Rev. Lett. **22**, 590 (1969).

7Y. C. Cheng, J. S. Clarke, and N. D. Mermin, Phys. Rev. Lett. **20**, 1486 (1968).

8G. A. Baraff, Phys. Rev. B **1**, 4307 (1970).

9G. A. Baraff, Phys. Rev. **187**, 851 (1969).

10See footnote 7 of Baraff, Grimes, and Platzman [Phys. Rev. Lett. **22**, 590 (1969)].

11See Eq. (28) of Mattis and Dresselhaus [Phys. Rev. **111**, 403 (1958)].

12See Fig. 2 of Mattis and Dresselhaus [Phys. Rev. Lett. **111**, 403 (1958)].

13E. Jensen and E. W. Plummer, Phys. Rev. Lett. **55**, 1919 (1985).

14A. W. Overhauser, Adv. Phys. **27**, 343 (1978).

15P. G. Coulter and W. R. Datars, Can. J. Phys. **63**, 159 (1985).

16F. E. Fragachán and A. W. Overhauser, Phys. Rev. B **29**, 2912 (1984).

17M. Huberman and A. W. Overhauser, Phys. Rev. B **25**, 2211 (1982).

18Xiaodong Zhu and A. W. Overhauser, Phys. Rev. B **33**, 925 (1986).

19A. W. Overhauser, Phys. Rev. Lett. **13**, 190 (1964).

20A. W. Overhauser, Phys. Rev. Lett. **55**, 1916 (1985).

21A. W. Overhauser and N. R. Butler, Phys. Rev. B **14**, 3371 (1976).

22P. A. Penz and T. Kushida, Phys. Rev. **176**, 804 (1968).

23G. F. Giuliani and A. W. Overhauser, Phys. Rev. B **20**, 1328 (1979).

24I. M. Lifshitz, M. Ya. Azbel, and M. I. Kaganov, Zh. Eksp. Teor. Fiz. **31**, 63 (1956) [Sov. Phys.—JETP **4**, 41 (1957)].

25G. E. H. Reuter and E. H. Sondheimer, Proc. R. Soc. London, Ser. A **195**, 336 (1948).

26E. A. Kaner and V. G. Skobov, Adv. Phys. **17**, 605 (1968).

27J. K. Galt, W. A. Yager, F. R. Merritt, B. B. Cetlin, and A. D. Brailsford, Phys. Rev. **114**, 1396 (1959).

28C. C. Grimes (private communication).

29Since the copper in the cavity was off the cylindrical axis, the $CuSO_4$ particles see mostly linearly polarized radiation. Accordingly, the resonances on either side of $H=0$ are of almost equal size.

30T. M. Giebultowicz, A. W. Overhauser, and S. A. Werner, Phys. Rev. Lett. (to be published).

31The spacing between the CDW gap and the first minigap is the same as that between the CDW gap and the Brillouin-zone gap (see Ref. 16).