Theory of Cyclotron Resonance Absorption by Negative-Mass Holes in Germanium

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The effect of orbital quantization on cyclotron resonance absorption by negative-mass heavy holes in germanium is considered. The Landau level system for the heavy holes in the approximation of an axial energy surface is computed with an equation derived by Firsov. A semiclassical technique is used to compute the cyclotron absorption by the holes in negative mass states under thermal equilibrium conditions. It is shown that at low temperatures the absorption is markedly less than that which would be calculated by the usual classical transport theory; at 4.2°K and 30 kMc/sec the absorption resonance essentially disappears. Similar effects are present, in principle, for any nonellipsoidal energy surface. The bearing of these calculations on several recent experiments is discussed.

RECENT interest in the negative-effective-mass heavy holes in germanium began with the work of Kroemer.[1] Dousmanis,[2] and at about the same time Firsov,[3] suggested the possibility of detecting a separate cyclotron resonance line for these holes by using circularly polarized microwaves. Subsequently, Dousmanis and co-workers[4] observed an emissive cyclotron resonance line which they attributed to light-excited negative-mass heavy holes. More recently, at a somewhat higher experimental frequency and under slightly different experimental conditions, Dexter *et al.*[5] observed no negative-mass resonance, emissive or absorptive. The properties of negative-mass carriers and the implications of the emissive result have been discussed by a number of authors.[6–13]

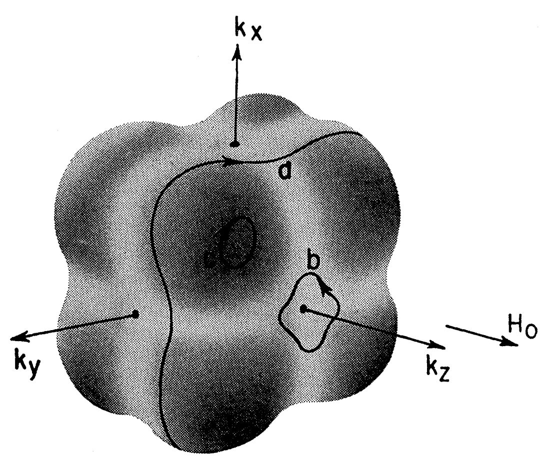

FIG. 1. A three-dimensional representation in **k** space of a surface of constant energy for the heavy holes in germanium. Typical hodographs are shown for the magnetic field in the k_z direction for: *a*, the usual positive-mass heavy holes; *b*, the negative-mass holes; and *c*, another type of positive-mass heavy hole. Arrows indicate the sense of carrier rotation.

In the present paper we consider the orbital quantization of the heavy holes in germanium in a magnetic field and use a semiclassical technique to calculate the cyclotron resonance absorption by the negative mass holes. The treatment yields a result distinctly different from that given by the usual classical Boltzmann treatment in that it shows a marked reduction in the negative-mass cyclotron resonance absorption at low carrier temperatures. This effect can remain large even for $\hbar\omega/kT<1$. While we treat only a specific case, such effects are present, in principle, for any nonellipsoidal energy surface.

INTRODUCTION

A collection of particles in an energy level system absorbs energy from a weak electromagnetic wave at a rate

$$\mathcal{P}(\omega)=\mathcal{P}_0\sum_{i,j} P_{ij}(\omega)(N_i-N_j),\quad \text{for}\quad E_i<E_j. \qquad (1)$$

Here N_i and N_j are the numbers of particles in the ith and jth levels, respectively, and E_i and E_j are their energies. $P_{ij}(\omega)$ $[=P_{ji}(\omega)]$ is the probability per

[1] H. Kroemer, Phys. Rev. **109**, 1856 (1958); Proc. Inst. Radio Engrs. **47**, 397 (1959); *Progress in Semiconductors*, edited by A. F. Gibson (Heywood and Company, Ltd., London, 1959), Vol. 4, p. 1; *International Solid-State Circuits Conference, 1960* (University of Pennsylvania Press, Philadelphia, Pennsylvania, 1960), p. 86.

[2] G. C. Dousmanis, Phys. Rev. Letters **1**, 55 (1958).

[3] Yu. A. Firsov, Fiz. Tverdogo Tela **1**, 44, 528 (1959) [Translations: Soviet Phys.—Solid State **1**, 42, 474 (1959)].

[4] G. C. Dousmanis, R. C. Duncan, J. J. Thomas, and R. C. Williams, Phys. Rev. Letters **1**, 404 (1958); Proceedings of the International Conference on Semiconductor Physics, Prague, 1960 [Czech. J. Phys. (to be published)]; G. C. Dousmanis, *Quantum Electronics*, edited by C. H. Townes (Columbia University Press, New York, 1960), p. 458.

[5] R. N. Dexter, D. Hensler, E. Hollar, and M. Halloran, Bull. Am. Phys. Soc. **5**, 177 (1960).

[6] C. Kittel, Proc. Nat'l. Acad. Sci. U. S. **45**, 744 (1959).

[7] D. T. Mattis and M. J. Stevenson, Phys. Rev. Letters **3**, 18 (1959).

[8] P. Kaus, Phys. Rev. Letters **3**, 20 (1959).

[9] S. Rodriguez, Phys. Rev. **115**, 821 (1959).

[10] R. C. Williams and F. Herman, Bull. Am. Phys. Soc. **5**, 61 (1960); Proceedings of the International Conference on Semiconductor Physics, Prague, 1960 [Czech. J. Phys. (to be published)].

[11] R. C. Duncan and B. Rosenblum, Bull. Am. Phys. Soc. **5**, 177 (1960); Proceedings of the International Conference on Semiconductor Physics, Prague, 1960 [Czech J. Phys. (to be published)].

[12] Yu. Kagan, Zhur. Ekspt'l i Teoret. Fiz. **38**, 1854 (1960) [Translation: Soviet Phys.—JETP **11**, 1333 (1960)].

[13] B. Lax, *Quantum Electronics*, edited by C. H. Townes (Columbia University Press, New York, 1960), p. 428.

particle that an incident photon of frequency ω induces a transition from i to j. Each P_{ij} includes a resonance denominator. It has been assumed that all of the levels are equally degenerate, as is the case for the Landau levels, and that the N_i are small compared to this degeneracy. The generalization is trivial.

Since the transition probabilities are positive definite, every term in the sum in Eq. (1) is positive if all $N_i > N_j$. Thus it is simply seen that the particles must *absorb* energy from the electromagnetic field if the population is everywhere a decreasing function of energy (a "normal population"). If, on the other hand, all $N_i < N_j$ (an "inverted population") the system must *emit* energy to the field. In a system which extends to infinite energy all N_j cannot be greater than all N_i if the total energy is to be finite. However, a "*net* inverted population" is still possible if there exists a region of population inversion within which the P_{ij} are sufficiently large compared to those in the region of the normal population. The particular nature of a negative mass system in no way alters these conditions for absorption or emission. In order for a negative-mass system to emit, net population inversion is required.

The behavior of carriers in electric and magnetic fields is determined by their dispersion law, i.e., the relation between their energy E and their wave vector $\mathbf{k}$. The dispersion law for the heavy holes of germanium[14] leads to constant energy surfaces in $\mathbf{k}$ space of the form shown three-dimensionally in Fig. 1. We will consider the case of an applied magnetic field $\mathbf{H}_0$ in the [001] direction. The classical motion of the holes in $\mathbf{k}$ space is then determined by the intersection of such energy surfaces with planes perpendicular to the magnetic field. Several such $\mathbf{k}$ space orbits (hodographs) are shown with arrows indicating the sense of rotation. From the form of the energy surfaces and the equation of motion, $\hbar d\mathbf{k}/dt = (e/\hbar c)\nabla_{\mathbf{k}}E \times \mathbf{H}_0$, it can readily be seen that $d\mathbf{k}/dt$ has opposite directions inside and outside the re-entrant regions near the [001] axis, i.e., the holes within the re-entrancy rotate in a sense opposite to that of those without. The frequency, sense of rotation, and cyclotron mass[15] for each hodograph are determined by the Shockley "tube-integral."[16] Hodographs such as that marked a correspond to the usual positive-mass heavy holes. Those lying within the re-entrant region, e.g., that marked b, correspond to negative cyclotron masses. Positive-mass hodographs of type c, which may also give rise to a distinct cyclotron resonance line or lines,[3,12] will be neglected by the approximations to be made below, but the bearing of the present work on such resonances will be briefly discussed. Since the orbits in real space have exactly the same shape and sense of rotation as those in $\mathbf{k}$ space, it is clear that the positive- and negative-mass holes interact principally with electric fields of opposite senses of circular polarization.

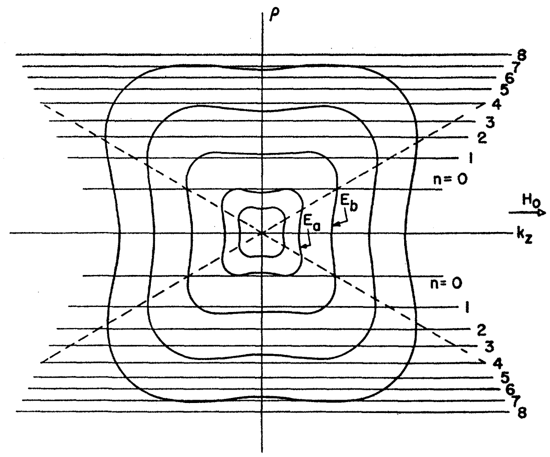

FIG. 2. Cross section of several surfaces of constant energy for the heavy holes in germanium in the axial approximation. The surfaces are figures of revolution about the k_z axis. The horizontal lines indicate the values of ρ allowed in the magnetic field H_0. The acute angle cones delineated by the broken lines are the regions of negative cyclotron mass. E_a is the lowest energy allowed; E_b is the lowest energy allowed for a negative-mass state.

Let us now consider the quantization of the energy of these carriers in a magnetic field. The energy level system for the actual germanium surfaces is extremely complicated. A considerable simplification is afforded by considering energy surfaces with axial symmetry about the magnetic field direction and ignoring the electron spin. While keeping the essential negative-cyclotron-mass features of the germanium surfaces, this approximation neglects the harmonics of the motion of the holes, resonances due to orbits such as those marked c in Fig. 1, and certain quantum effects which are important only when $(\hbar\omega/kT) \gtrsim 1$. None of these are likely to be significant for present considerations.

The cross section of a family of such axial approximations to the germanium surfaces is shown in Fig. 2. The surfaces are figures of revolution about the k_z axis. The acute angle cones delineated by the dotted lines are the regions of negative cyclotron mass.

Quantization of the angular momentum of the particles in a magnetic field by the Bohr-Sommerfeld quantization rule yields allowed hodographs lying on a family of coaxial cylinders about the k_z axis as shown in Fig. 2. The radii of the allowed hodographs are given by

$$\rho = (k_x^2 + k_y^2)^{\frac{1}{2}} = [(2eH_0/\hbar c)(n+\tfrac{1}{2})]^{\frac{1}{2}}, \qquad (2)$$

where n is the Landau level quantum number. From this result it can readily be shown that the $\mathbf{k}$ space volume of each cylindrical shell is the same. The same number of $\mathbf{k}$ space states have "coalesced" to each cylinder, and the Landau levels are thus equally

[14] G. Dresselhaus, A. F. Kip, and C. Kittel, Phys. Rev. **98**, 368 (1955).

[15] The usual transverse mass, $(d^2E/\hbar^2 dk_t^2)^{-1}$, need not be considered. It is possible for particles with negative cyclotron mass to have positive transverse mass.

[16] W. Shockley, Phys. Rev. **79**, 191 (1950).

degenerate. For cyclotron resonance, where the microwave electric field is perpendicular to $\mathbf{H}_0$, the resonant transitions are vertical in Fig. 2, i.e., $\Delta k_z=0$. Furthermore, the correspondence principle implies the selection rule $\Delta n=\pm 1$. In regions of positive cyclotron mass, where energy increases with n, $\Delta n=+1$ corresponds to a power absorbing transition, while in regions of negative cyclotron mass energy decreases with increasing n, and $\Delta n=-1$ corresponds to absorption.

In the next section we will calculate the cyclotron resonance absorption by the negative-mass holes for an axial approximation to the energy surfaces of germanium. An important feature of the result of this calculation can be seen qualitatively from a study of Fig. 2. The quantization prevents carriers from occupying energy surfaces with energy less than E_a. Those carriers with energies between E_a and E_b can only be in positive-mass regions of $\mathbf{k}$ space. Only at energies above E_b can carriers occupy negative-mass states. Therefore, if the carrier distribution temperature is sufficiently low, i.e., $kT \ll E_b-E_a$, an extremely small fraction of the carriers will occupy the negative-mass region. If, on the other hand, $kT \gg E_b-E_a$ positive- and negative-mass states are occupied in proportion to their respective classical volumes in $\mathbf{k}$ space. In the latter case, the ratio of negative-mass cyclotron resonance absorption to positive-mass absorption would be that calculated classically. However, if $kT \ll E_b-E_a$ this ratio becomes vanishingly small. This is a consequence of the quantization and, of course, is not given by the classical theory. Furthermore, since $(E_b-E_a)/\hbar\omega_c$, where $\omega_c=eH_0/m^*c$, depends on the extent of the warping of the energy surfaces, kT being large compared to $\hbar\omega_c$ (the level spacing) does not in itself ensure the validity of the classical result. In fact, as will be shown, this orbital quantization effect is of considerable importance for the negative-mass holes in germanium under the usual cyclotron resonance experimental conditions.

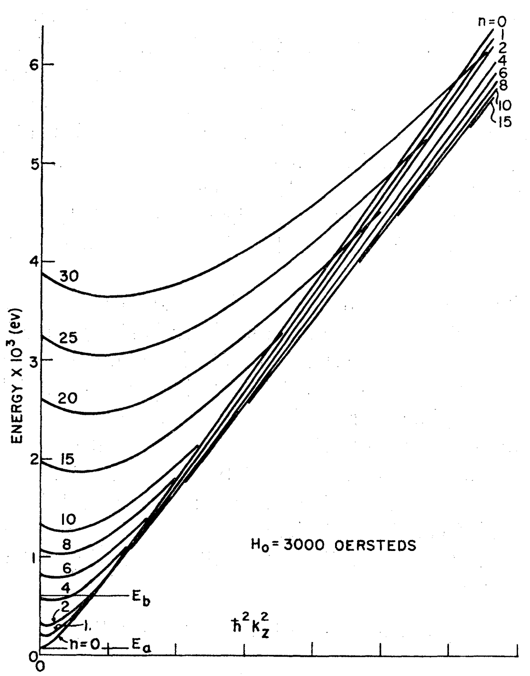

FIG. 3. Landau level energies for the heavy holes in germanium in the axial approximation as a function of the square of the momentum in the magnetic field direction. When, for a fixed k_z, energy decreases with increasing quantum number n, the levels correspond to negative-mass states. For clarity, only certain representative levels are plotted.

CALCULATION OF THE NEGATIVE MASS CYCLOTRON RESONANCE ABSORPTION

The equation of the surfaces of constant energy for the heavy holes in germanium can be written

$$E=A(\rho^2+k_z^2)-[B^2(\rho^2+k_z^2)^2+C^2(k_z^2\rho^2+k_x^2k_y^2)]^{\frac{1}{2}}, \quad (3)$$

where A, B, and C are constants.[14] The allowed energies of these holes in the presence of an external magnetic field have been calculated for those carriers with $k_H=0$, where k_H $(\equiv k_z)$ is the component of $\mathbf{k}$ in the magnetic field direction,[17] and for carriers with $k_H\neq 0$ in the approximation of spherical energy surfaces.[18] Unfortunately, the negative-mass feature we wish to consider does not exist in these approximations.[19] However, for carriers without spin and with energy surfaces having axial symmetry about the magnetic field direction, the exact quantum mechanical expression for the allowed energy as a function of k_z and n has been given by Firsov.[3] Such axial surfaces can exhibit the essential negative mass features of the germanium surfaces; the features which are neglected (see above) are of minor importance for the present calculation. Their omission is warranted by the considerable simplification afforded and in view of other approximations which will be made.

We, therefore, average out the azimuthal dependence of Eq. (3). This can be done to a sufficient degree of approximation simply by neglecting the azimuthal dependence of ρ, which is quite small, and replacing the non-axially-symmetric term in Eq. (3) by its average,

$$\langle k_x^2k_y^2\rangle_{\mathrm{av}}=\frac{1}{2\pi}\int_0^{2\pi} k_x^2k_y^2 d\phi=\tfrac{1}{8}\rho^4. \quad (4)$$

With this approximation Eq. (3) becomes

$$E=A(\rho^2+k_z^2)-[(B^2+\tfrac{1}{8}C^2)\rho^4+B^2k_z^4 + (2B^2+C^2)k_z^2\rho^2]^{\frac{1}{2}}. \quad (5)$$

The general solution given by Firsov for the Landau

[17] W. Kohn and J. M. Luttinger, Phys. Rev. **96**, 529 (1954); J. M. Luttinger and W. Kohn, *ibid.* **97**, 869 (1955); J. M. Luttinger, *ibid.* **102**, 1030 (1956).

[18] R. F. Wallis and H. J. Bowlden, Phys. Rev. **118**, 456 (1960).

[19] See also V. Evtuhov, California Institute of Technology, Electron Tube and Microwave Laboratory, Technical Report No. 16, 1960 (unpublished). This recent treatment is somewhat more general and includes results for $k_H\neq 0$ with non-spherical energy surfaces.

level system for carriers with such energy surfaces is[20]

$$E_n(k_z)\equiv E_{n,k}=(eH_0/\hbar c)2A\,(n+\tfrac{1}{2})+(eH_0/\hbar c)\alpha+Ak_z^2 -\{[(eH_0/\hbar c)\beta(n+\tfrac{1}{2})+(eH_0/\hbar c)\alpha+Bk_z^2]^2 +(eH_0/\hbar c)\gamma(n+1)k_z^2\}^{\frac{1}{2}}, \quad (6)$$

where

$$\alpha=A+(B^2+\tfrac{1}{8}C^2)^{\frac{1}{2}},$$
$$\beta=2(B^2+\tfrac{1}{8}C^2)^{\frac{1}{2}},$$
$$\gamma=4[B^2+\tfrac{1}{2}C^2-B(B^2+\tfrac{1}{8}C^2)^{\frac{1}{2}}],$$

and where

$$A=13.1\ (\hbar^2/2m_0),\quad B=8.3\ (\hbar^2/2m_0),\quad C=12.5\ (\hbar^2/2m_0)$$

are the values used for the constants of Eq. (3).[21]

We have computed the Landau level system given by Eq. (6). The numerical evaluation was done with the IBM 650. In Fig. 3 the energy for several representative values of n is plotted as a function of $\hbar^2k_z^2$ for a magnetic field of 3000 oe. This is approximately the magnetic field at which the negative-mass holes near the [001] axis resonate at a frequency of 30 kMc/sec. For a fixed k_z, those levels for which energy decreases with increasing quantum number correspond to negative-mass states. The energy at which the first such state occurs is about 8×10^{-4} ev for this magnetic field and increases with increasing magnetic field. The general form of Fig. 3 could readily be deduced from Fig. 2.

We will now calculate the power absorbed by a distribution of carriers in this level system by the use of an appropriate form of Eq. (1). Since we have the selection rule $\Delta k_z=0$, and since there is essentially no quantization of k_z, the sum in Eq. (1) can conveniently be replaced by a sum over all states in a k_z plane and an integral over k_z.

For isotropic positive masses in a parabolic band, the transition probability between states l and n is nonzero only if $|n-l|=1$; and since $E_{n,k}<E_{n+1,k}$, we can write Eq. (1) as

$$\mathcal{P}^+=\text{const}\times\int_{-\infty}^{\infty}dk_z\sum_{n=0}^{\infty}(N_{n,k}-N_{n+1,k})P_{n,n+1}. \quad (7)$$

It can be shown that for this case

$$\mathcal{P}^+=\int_{-\infty}^{\infty}dk_z\sum_{n=0}^{\infty}(N_{n,k}-N_{n+1,k})\frac{e^2\tau}{|m^*|}\frac{E_0^2}{2}\times\left[\frac{1}{1+(\omega-\omega_c)^2\tau^2}+\frac{1}{1+(\omega+\omega_c)^2\tau^2}\right](n+1), \quad (8)$$

where E_0 is the amplitude of the linearly polarized microwave electric field, and τ is the carrier momentum relaxation time. This is essentially the equation derived by Argyres.[22]

If the effective mass is negative, then $E_{n+1,k}<E_{n,k}$, and the condition $E_i<E_j$ on the summation in Eq. (1) forces us to interchange $N_{n,k}$ and $N_{n+1,k}$ in Eqs. (7) and (8). The $P_{n,n+1}$ $(=P_{n+1,n})$ have exactly the same form as in the positive-mass case. Only the absolute value of m^* enters since the transition probability is determined by the square of the absolute value of the matrix element. Actually, one may write Eq. (8) for both the positive- and negative-mass cases by replacing $|m^*|$ with m^*. The interpretation of the coefficient of the population difference as a transition probability must then be relaxed.

The treatment of a level system where the level spacing (i.e., the effective mass) is a function of $\mathbf{k}$ would be considerably more complicated. If one can assume that the fractional change in level spacing is small over a region of $\mathbf{k}$ space containing a large number of levels, a considerable simplification is possible in the calculation of the power absorption. It is then permissible to use in Eq. (8) the transition probability for the constant mass case with the appropriate values of m^* and ω_c for each region of $\mathbf{k}$ space, i.e., $m^*=m^*(n,k_z)$ and $\omega_c=\omega_c(n,k_z)\equiv\omega_{n,k}$. This is the semiclassical approximation.

Equation (8) can readily be transformed into the classical expression[23] for the power absorbed by carriers in a band of arbitrary dispersion law by changing the sum to an integral and making the substitutions

$$n=\rho^2(\hbar c/2eH_0)\gg1,\quad N_{n,k}=f(\mathbf{p})(2eH_0/\hbar^2c), \quad (9)$$

where $f(\mathbf{p})$ is the classical distribution function and $\mathbf{p}=\hbar\mathbf{k}$.

We will use the semiclassical approximation and Eq. (8) to calculate the power absorbed by the negative-mass holes in germanium under thermal equilibrium conditions. The fact that some of the negative-mass levels shown in Fig. 3 do not meet the requirements for the validity of the approximation will preclude a high degree of quantitative accuracy in calculations for very low temperatures. Such inaccuracies are not particularly significant in the present calculation, which estimates the size of a rather gross effect. The major part of the positive-mass energy level system is composed of approximately equally spaced levels even at the lower energies. Therefore the absorption by the positive-mass carriers can be calculated quite validly on a purely classical basis as long as $\hbar\omega$ is moderately small compared to kT.

For a Boltzmann distribution

$$N_{n,k}=(N/Z)\exp[-E_{n,k}/kT],$$

[20] We are indebted to Yu. A. Firsov for confirming the existence of two typographical errors in his Eq. (6) of reference 3. The factor 4 multiplying (n+1) in the last term of that equation should be omitted and the accompanying definition of $\omega_{1,2}$ should contain another factor of $\hbar$ in the denominator.

[21] B. Lax, Revs. Modern Phys. 30, 122 (1958).

[22] P. Argyres, Phys. Rev. 109, 1115 (1958).

[23] A particularly convenient form is given by Williams and Herman (see reference 10).

where N is the total number of carriers in the band (both positive and negative mass) and

$$Z=\int_{-\infty}^{\infty} dk_z \sum_{i=0}^{\infty} \exp-(E_{i,k}/kT) \qquad (10)$$

is the partition function. In order to evaluate the partition function we need only consider the contribution made to it by the positive mass states since they constitute about 90% of $\mathbf{k}$ space. Furthermore, when $\hbar\omega$ is moderately small compared to kT, the positive-mass level system can be closely approximated by that of free particles in a magnetic field with the average effective mass $\bar{m}$ of the heavy holes. Therefore

$$Z=\int_{-\infty}^{\infty} dk_z \sum_{n=0}^{\infty} \exp\left[-\frac{(n+\frac{1}{2})e\hbar H_0}{\bar{m}ckT}-\frac{\hbar^2k_z^2}{2\bar{m}kT}\right]$$
$$\approx(2\pi\bar{m}kT)^{\frac{1}{2}}\left[\frac{\bar{m}kTc}{e\hbar^2H_0}\right]. \qquad (11)$$

Since the spacing between the energy levels we consider is moderately small compared to kT, we will approximate $N_{n+1,k}-N_{n,k}$ by $(dN_{n,k}/dE_{n,k})dE_{n,k}$. Thus

$$N_{n+1,k}-N_{n,k}=-\frac{N}{ZkT}\exp-(E_{n,k}/kT)\hbar\omega_{n,k}, \qquad (12)$$

where $\hbar\omega_{n,k}=E_{n+1,k}-E_{n,k}$. With these approximations Eq. (8) becomes

$$\mathcal{P}=\frac{-2\pi e^2E_0^2N\hbar^3\tau}{(2\pi\bar{m}kT)^{\frac{3}{2}}kT}\times\int_{-\infty}^{\infty} dk_z \sum_{n=0}^{\infty}\frac{\exp-(E_{n,k}/kT)\omega_{n,k}^2(n+1)}{1+(\omega+\omega_{n,k})^2\tau^2}, \qquad (13)$$

where we have replaced m^* by $eH_0/c\omega_{n,k}$ and assumed that the incident microwave field is circularly polarized with the proper polarization to interact with the negative masses.

With this equation the cyclotron resonance absorption by the negative-mass holes was computed as a function of H_0 for two frequencies, 15 and 30 kMc/sec, and two carrier distribution temperatures, 50 and 4.2°K. It was assumed that the total number of holes in the valence band did not change with the carrier distribution temperature. (Or alternatively the absorption is normalized to a fixed number of carriers.) We have taken τ as constant throughout $\mathbf{k}$ space and equal to 5×10^{-11} second in each case. For each value of H_0 for which a point was computed the entire level system (i.e., the $E_{n,k}$) had to be redetermined from Eq. (6) and the $\omega_{n,k}$ evaluated. The sum indicated in Eq. (13) could then be performed for a series of values of k_z. The sum was stopped at that $n=m$ for which the $E_{n,k}$ begin to increase with increasing n. Thus the absorption by only those carriers in negative mass levels was included. The integral was approximated by appropriately weighting these sums and adding them. An IBM 650 computer was used for the numerical evaluation. The absorption was not computed at very low H_0 since the number of levels which must be included becomes large and the computer time required increases rapidly. When $H_0=0$ the absorption can be calculated classically and, of course, is independent of temperature. The results are plotted in Fig. 4.

The absorption shown is only that due to the negative-mass holes. Since we consider circularly polarized microwaves, the absorption peak of positive-mass holes would appear for the magnetic field in the opposite direction. This peak absorption would be several hundred times the largest in Fig. 4. Experimentally, the absorption shown would be added to the tail of this positive-mass resonance. The width of the resonances shown is determined by the large spread in the values of the negative masses; the positive-mass resonance is considerably sharper for the value of τ we have chosen.

DISCUSSION

The decrease in strength of the negative-mass absorption when the carrier temperature is lowered to 4.2°K is considerable at 15 kMc/sec and striking at 30 kMc/sec. The reasons for this decrease, already discussed in the Introduction in terms of Fig. 2, can now, perhaps, be seen more clearly by consideration of the calculated energy level system shown in Fig. 3. A 4.2°K Boltzmann distribution ($kT=0.36\times10^{-3}$ ev) populates relatively few negative-mass levels and these largely in the region where they are not equally spaced. Therefore a weak and poorly defined negative-mass resonance is expected. At 50°K ($kT=4.3\times10^{-3}$ ev), on the other hand, many equally spaced levels are populated, and a stronger more sharply defined resonance results. The displacement of the 4.2°K absorptions to lower magnetic field is a result of the negative mass levels disappearing from the low-energy region with increasing magnetic field faster than the spacing between them becomes resonant. A classical calculation, i.e., one which neglects the orbital quantization, would show no such temperature dependence. The curve for 15 kMc/sec and 50°K is not far from the classical case. The value of $\hbar\omega/kT$ for which these effects become important is not necessarily in the region of unity, but depends on the extent of the negative mass region. The smaller the negative-mass cone angle the smaller that value of $\hbar\omega/kT$ at which the absorption decreases. (For the curves of Fig. 4, $\hbar\omega/kT$ is, in order of decreasing amplitude, 0.014, 0.029, 0.17, and 0.34.)

The predicted temperature dependence of the intensity of the negative mass resonance could be checked by an experiment in which a suitably doped sample of germanium (or silicon) is warmed above helium temperature. Above about 10°K a sufficient number of

acceptors could be ionized to provide a detectable signal with the carrier distribution in thermal equilibrium. The quantitative agreement between the results of such an experiment and the present calculation would, of course, be limited by the approximations used. The experiment at 24 kMc/sec with light excitation reported by Dexter *et al.*[5] showed no negative-mass resonance. This is the result which the present calculation would predict if their holes were thermalized. Their sensitivity was probably high enough that they would have observed a resonance if it were there with its classically predicted strength.

In an experiment where one heats the carrier distribution, carrier energy effects of the type discussed would also be expected from the regions of the energy surface other than the re-entrance, i.e., the protrusions in the [111] directions which give rise to hodographs such as c in Fig. 1. In a recent paper, Levinger and Frankl,[24] using a microwave power large enough to heat the carrier distribution, observed distortions on the heavy-hole resonance. These may in part be due to such effects.

Since the present paper was stimulated, as were most of the recent publications on negative masses, by the observation of an emissive negative-mass cyclotron resonance by Dousmanis and co-workers, some comment is probably in order concerning the relationship of the effects we have just described to this experiment. The emissive resonance must result from a net population inversion in the negative-mass region. The present paper does not treat the problem of how such a net population inversion might come about. However, the *reduced absorption* at low temperature calculated above may have some bearing on the emissive result.

Under the conditions of the experiment of Dousmanis *et al.*, no carriers are excited to the very high-energy regions (for holes) of the valence band, and the population of the negative mass states cannot be inverted over the entire band. Therefore, the existence of a net inversion would require the transition probability to be small at the higher energies where the population is not inverted. (A possible reason for a small transition probability is the short phonon emission time for hot carriers.) The time required for the light-excited holes to come to quasi-thermal equilibrium in the valence band (of the order of the phonon emission time) is almost certainly much shorter than the recombination time for these holes. Therefore, in the steady state the holes are approximately in a thermal distribution at the lattice temperature. However, that small fraction recently excited by the light and yet unthermalized could conceivably have an inverted distribution. In a classical calculation, where the fraction of holes in the negative-mass states does not depend on hole energy, absorption by the thermalized negative-mass carriers

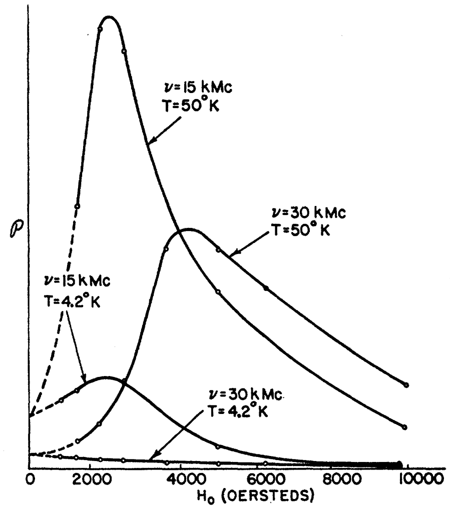

FIG. 4. Computed cyclotron resonance absorption for negative-mass heavy holes in an equilibrium distribution in germanium at two temperatures, 4.2°K and 50°K, and two frequencies, 15 kMc/sec and 30 kMc/sec, vs magnetic field in the [001] direction. Circles indicate computed points. The broken lines indicate the extrapolation to zero magnetic field, where the absorption must be independent of temperature.

could outweigh any emission by the few at higher energies and thus yield a net absorption. Due to the orbital quantization, however, the cyclotron absorption at the negative-mass resonance by holes thermalized at low temperatures is very small. Thus a small inversion at higher energies (for which we offer no explanation) could possibly produce a net emission at the negative-mass resonance in spite of the fact that the total distribution function may not be far from a Boltzmann distribution. In such a case, the positive-mass resonance, dominated by the large number of thermalized carriers, would still be strongly absorptive. In order to explain an emissive negative-mass resonance and a simultaneously absorptive positive-mass resonance without considering the effects of quantization, one must assume an anisotropy of the distribution function in momentum space.[25] The quantization allows this other interpretation in which the distribution function may be isotropic (essentially by introducing an anisotropy in the density-of-states function at low energies).

The emissive anisotropic distributions of Williams and Herman[10] (and also that originally suggested by Kroemer[1]), in which states near the k_z axis are preferentially populated, are, of course, population inversions in the sense of Eq. (1). This can be seen by correlating the Landau levels in Fig. 3 with appropriate angles in $\mathbf{k}$ space, i.e., by comparing Fig. 2 and Fig. 3. Anisotropic distributions are of particular interest since they may,

[24] B. Levinger and D. Frankl, Phys. Rev. Letters 5, 12 (1960).

[25] Williams and Herman (reference 10) have made extensive calculations of the effects of distributions containing anisotropy.

perhaps, be artificially created.[1,26] Since the thermalized portion of any distribution is isotropic, the importance of orbital quantization considerations is essentially the same for isotropic and anisotropic distributions.

We offer no explanation for the difference between the experimental results of Dousmanis *et al.* and Dexter *et al.*, except, perhaps, to point out that, due to the quantization, a region of energy allowed for negative-mass carriers in the former experiment is not allowed at the higher magnetic fields of the latter.

[26] If the strongly anisotropic distribution envisioned in reference 1 were actually achieved, the emissive effect would be enhanced by a factor of about $(1+\omega^2\tau^2)$ by the application of a magnetic field to resonate the negative masses.

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Internal Fields at Low Temperatures in CoPd Alloys*

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The hyperfine splitting of the 14.4-kev gamma kev ray line in Fe^{57} has been measured for a series of sources, each containing Co^{57} activity doped into a host lattice of CoPd. Although Pd itself is not ferromagnetic, the alloys with Co are all ferromagnetic, with Curie temperatures ranging from 1404°K for pure Co down to 130°K for a 3% Co alloy (the most dilute which we have studied). The internal field associated with the hyperfine splitting is a function of temperature for a given alloy; however, at temperatures small compared to the Curie temperature, we find that each source shows very nearly the same internal field, namely −308 kgauss. The relationship of this behavior to current theories of the internal field in Fe and to the nature of ferromagnetism in CoPd is briefly discussed.

THE ferromagnetism of alloys of Pd with the iron series elements has been extensively studied.[1–3] The Co-Pd alloys are a continuous series of random substitutional solid solutions.[4] The dilute Co alloys at room temperature are fcc in structure and show no superlattice structure.[4] They have the interesting property that, although Pd itself is not ferromagnetic, the addition of as little as 0.1% Co makes an alloy which is ferromagnetic.[1] This raises the question of the mechanism for this very long range ferromagnetism.

The experimental information presently available for the cobalt-palladium alloys concerns bulk properties such as the Curie temperature T_c, the saturation magnetization, and the magnetic susceptibility. Recently, however, the Mössbauer effect has been used to study the internal magnetic field H_i at Fe^{57} impurities in various materials. For Fe^{57} in a lattice[2] of pure Fe, we have reported the temperature dependence of the hyperfine splitting.[5] The absorption spectrum at all temperatures from 1026° to 0.35°K showed six well-resolved lines, indicating H_i must be very nearly the same at all lattice positions. (The Curie temperature of Fe is 1046°K.) Furthermore, H_i and the saturation magnetization have the same temperature dependence, within the accuracy of the experiment. For the Co-Pd alloys, because of their random character, it seemed interesting to investigate with the Mössbauer effect whether H_i would still be unique, and what would be the temperature dependence.

A series of Co-Pd alloys were prepared ranging in Co concentrations from 100% to 3% Co. Each sample was rolled to about 5 mils thickness and electroplated with Co^{57}, which was then diffused into the lattice for 1 hour at 1000°C. The Mössbauer effect spectrum was obtained at several temperatures for each alloy. Some of the 8% samples in the interval $T_c > T > 0.7\ T_c$ show anomalous spectra, which appear to be the superposition of a six-line hyperfine pattern and a single-line (unsplit) pattern. This effect is still being studied. However, for $T < 0.5\ T_c$, every sample showed a spectrum of six well-defined emission lines, indicating that H_i was very nearly the same at all lattice positions for a given sample. From the splitting, we obtain H_i in kilogauss,

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[1] R. M. Bozorth, P. A. Wolff, D. D. Davis, V. B. Compton, and J. H. Wernick, Phys. Rev. **122**, 1157 (1961).
[2] E. O. Wollan, Phys. Rev. **122**, 1710 (1961).
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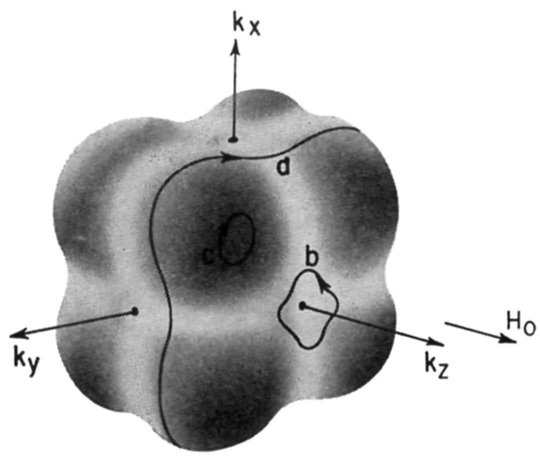

FIG. 1. A three-dimensional representation in **k** space of a surface of constant energy for the heavy holes in germanium. Typical hodographs are shown for the magnetic field in the k_z direction for: a, the usual positive-mass heavy holes; b, the negative-mass holes; and c, another type of positive-mass heavy hole. Arrows indicate the sense of carrier rotation.