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Ultrasonic Amplification in Semimetals

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When a semimetal is placed in crossed electric (**E**) and magnetic fields (**H**), both electrons and holes drift in the $\mathbf{E}\times\mathbf{H}$ direction with velocity Ec/H. A sound wave propagating in this direction with velocity s may be amplified if $Ec/H>s$. The effect is similar to that described by Hutson, McFee, and White, except that the present arrangement for semimetals allows one to obtain conditions of amplification at much lower current densities, since the magnetic field limits the current flowing in the electric field direction. The theory of sound amplification in a semimetal has been worked out for the above conditions, assuming the field produced by the sound wave can be described by a deformation potential. Because of the relatively large carrier densities in semimetals such as bismuth, it is found that large amplification factors can be obtained.

INTRODUCTION

Recently, Hutson, McFee, and White[1] have shown that ultrasonic amplification is possible in CdS crystals, when the drift velocity of optically excited electrons in an electric field is greater than the velocity of sound. Substantial acoustic gain is observed in CdS even though the number of free carriers is relatively small, because of the strong piezoelectric interaction between the carriers and long-wavelength ultrasonic waves. In principle, there is no reason why the above mentioned effect could not also be observed in materials which are not piezoelectric,[2] but which have higher carrier concentrations. The chief practical difficulty in these cases arises from the fact that by the time the carriers have drift velocities comparable to the velocity of sound, the current density is very large. For example, in bismuth, a drift velocity of 10^5 cm/sec implies a current density of about 6×10^3 amp/cm^2 at 2°K, whereas for materials with higher carrier concentrations the corresponding current densities are even higher. In the present paper, we offer an alternate arrangement, suggested by a recent experiment of Esaki,[3] appropriate for observing ultrasonic amplification in semimetals, which largely overcomes the above-mentioned practical difficultly. The new scheme employs perpendicular electric **E** and magnetic fields **H**. With this arrangement, both electrons and holes drift in the $\mathbf{E}\times\mathbf{H}$ direction with velocities Ec/H (c=velocity of light). We shall presently show that an ultrasonic wave propagating with velocity s in the $\mathbf{E}\times\mathbf{H}$ direction may be amplified by the current carriers when $Ec/H>s$. Although there is a qualitative similarity between our scheme and that used by Hutson *et al*,[1] there are important differences between these approaches. First of all, when $Ec/H>s$ (a necessary condition for amplification), the drift velocity *in the direction of the electric* field is of the order $s/\omega_c\tau$, where ω_c is the cyclotron frequency and τ is the scattering time. The current densities required for amplification are, therefore, smaller than those required in the technique of Hutson *et al.*[1] by a factor $1/\omega_c\tau$. In bismuth, for example, where $\omega_c\tau\simeq10^3$ is attainable at low temperatures, the required current density is 6 amp/cm^2 instead of 6×10^3 amp/cm^2. On the other hand, the power densities required for the onset of amplification are the same in the two methods—approximately 100 watt/cm^3 for bismuth at low temperatures. Nevertheless, the application of a magnetic field raises the impedance level of the sample and thereby reduces contact and series resistance problems.

Essential to the present technique is the fact that no appreciable Hall voltage is set up for materials with equal electron and hole concentrations, when the applied magnetic field is large. A similar effect should be observable for materials with just one type of carrier, in arrangements where no Hall field is allowed to build up (e.g., Corbino disk).

From the theoretical point of view, the present technique should help to clarify the band structure of semimetals by making it possible to study ultrasonic effects such as cyclotron resonance and geometrical resonance[4] under conditions of *amplification* rather than attenuation. Resonance effects will appear, as they do in the absence of an electric field, when the cyclotron orbit radius r_c or frequency ω_c is comparable to the wavelength λ, or frequency ω, of the ultrasonic wave.[4] However, when an electric field is present, the effective frequency, ω_{eff}, of the wave is

$$\omega_{\text{eff}}=\omega\left[1-\left|\frac{Ec/H}{s}\right|\right],$$

because of the carrier drift velocity. In this paper we shall present the theory of ultrasonic amplification in the presence of crossed electric and magnetic fields using the dc expressions for the carrier mobility and diffusion constants. Such a treatment is valid for $\lambda\gg r_c$ and $\omega_{\text{eff}}\ll\omega_c$. Extension of the present theory to resonance conditions will be the subject of a future publication.

We have mentioned above, that according to the theory to be presented, an ultrasonic wave traveling

[1] A. R. Hutson, J. H. McFee, and D. L. White, Phys. Rev. Letters **7**, 237 (1961).
[2] G. Weinreich, Phys. Rev. **104**, 321 (1956).
[3] L. Esaki, Phys. Rev. Letters **8**, 4 (1962).
[4] M. H. Cohen, M. J. Harrison, and W. A. Harrison, Phys. Rev. **117**, 937 (1960).

in the $\mathbf{E}\times\mathbf{H}$ direction will be attenuated by the carriers when $Ec/H<s$. Since under this condition the carriers are continuously taking energy from the wave, it is clear that our theory can apply in a strict sense only to vibrational modes of large amplitude, i.e., modes which are not in thermal equilibrium with the rest of the crystal. In a qualitative sense, however, the theory may also be used to discuss amplification in a thermal ensemble of phonons. If we consider an increase in the number of phonons in a mode due to statistical fluctuations, the electron-phonon interaction will tend to reduce this number towards the average value of $kT/\hbar\omega$ if $Ec/H<s$. However, if $Ec/H>s$, the electrons will cause the fluctuation to grow until some new effect (anharmonic effects, nonlinear recombination, etc.) prevents further growth. This qualitatively describes the effect observed by Hutson *et al.*[1] when no input signal is used. We believe that the recent observations by Esaki[3] of a discontinuity of the conductivity of bismuth, when $Ec/H\simeq s$, is also related to this "heating up" of certain modes of vibration.

CALCULATION OF ATTENUATION CONSTANT

In calculating the average rate of energy transfer per unit volume $\langle dW/dt\rangle$, we shall represent the effects of the sound wave on the electrons, n, and holes, p, by deformation potentials. We will consider a dc electric field $\mathbf{E}$ to be applied in the x direction, and a constant magnetic field $\mathbf{H}$ applied in the $-z$ direction. Under these conditions, electrons and holes will drift in the y direction with velocity Ec/H (assuming fields H, such that $\omega_c\tau\gg1$). The electron and hole deformation potentials V_n and V_p due to an acoustic wave of wave number k and frequency ω traveling in the y direction may then be written:

$$V_n(y,t)=e\mathcal{E}_n\epsilon\exp i(ky-\omega t), \tag{1a}$$

$$V_p(y,t)=e\mathcal{E}_p\epsilon\exp i(ky-\omega t), \tag{1b}$$

where $\mathcal{E}_n$ and $\mathcal{E}_p$ are deformation potential constants, measured in volts, for electrons and holes, respectively, and ϵ is the strain accompanying the wave. Both $\mathcal{E}_n$ and $\mathcal{E}_p$ will depend in general upon the directions of propagation and polarization of the wave, but since we are dealing with only one mode, we have chosen to use the simplified notation of Eq. (1). The effective electric fields acting on the electrons and holes as a result of the acoustic wave will be

$$E_n=\mathcal{E}_n\epsilon k e^{i(ky-\omega t+\frac{1}{2}\pi)}, \tag{2a}$$

$$E_p=\mathcal{E}_p\epsilon k e^{i(ky-\omega t+\frac{1}{2}\pi)}. \tag{2b}$$

In order to simplify the calculation of the electron and hole currents, we shall use an approximation commonly used in dealing with space-charge effects in materials of high carrier density. We shall represent the deviations from space-charge neutrality as giving rise to an additional electric field $E_s(y,t)$ but apart from this, we will treat the crystal as being space-charge neutral ($n=p$). The validity of this approximation may later be verified, insofar that it is possible to calculate how large a deviation from space-charge neutrality would be required to maintain the field E_s. The electron and hole current densities in the y direction are then

$$j_n=-e\left[n\frac{Ec}{H}-D_n\frac{dn}{dy}-n\mu_n(E_n+E_s)\right], \tag{3a}$$

$$j_p=e\left[n\frac{Ec}{H}-D_p\frac{dn}{dy}+n\mu_p(E_p+E_s)\right]. \tag{3b}$$

In Eqs. (3a) and (3b), D_n, D_p are diffusion constants and μ_n, μ_p are carrier mobilities, all in a transverse magnetic field. As discussed in the introduction, we shall represent these quantities by their values for $k=0$ and $\omega_{\text{eff}}=0$. We then have

$$\mu_n=\frac{e\tau_n}{m_n}\frac{1}{(\omega_{cn}\tau_n)^2},\qquad \mu_p=\frac{e\tau_p}{m_p}\frac{1}{(\omega_{cp}\tau_p)^2}, \tag{4a}$$

$$eD_n=\tfrac{2}{3}\zeta_n\mu_n,\qquad eD_p=\tfrac{2}{3}\zeta_p\mu_p, \tag{4b}$$

where τ_n is the electron scattering time, m_n the electron effective mass, ζ_n the electron Fermi level, and ω_{cn} is the electron cyclotron frequency. Similar definitions are valid for the holes. We shall assume that conditions at the surfaces of the sample are such that concentration gradients in the y direction may be ignored. The continuity equations for electrons and holes are then,

$$e\frac{dn}{dt}=-e\frac{n-n_0}{\tau_r}+\frac{dj_n}{dy}, \tag{5a}$$

$$e\frac{dn}{dt}=-e\frac{n-n_0}{\tau_r}-\frac{dj_p}{dy}, \tag{5b}$$

where n_0 is the equilibrium electron (or hole) concentration and τ_r is the recombination time. Multiplying Eq. (5a) by μ_p and Eq. (5b) by μ_n and adding we obtain an equation for n independent of the space-charge field E_s:

$$\frac{dn}{dt}=-\frac{n-n_0}{\tau_r}-\frac{Ec}{H}\frac{dn}{dy}+D\frac{d^2n}{dy^2}+\mu[E_n-E_p]\frac{dn}{dy}+n\mu\frac{d}{dy}[E_n-E_p], \tag{6}$$

where

$$D=[\mu_pD_n+\mu_nD_p]/(\mu_n+\mu_p);\quad \frac{1}{\mu}=\frac{1}{\mu_n}+\frac{1}{\mu_p}.$$

Once Eq. (6) is solved for n, the space-charge field $E_s(y,t)$ may be determined from the relation $j_n+j_p=0$. In order to solve Eq. (6), we replace n by n_0 in the last term and ignore the term $\mu(E_n-E_p)dn/dy$, since it is

small compared to the last term if we make the reasonable assumption that $n-n_0 \ll n_0$. Then putting $n-n_0 = Ae^{i[ky-\omega t+\frac{1}{2}\pi]}$, we find

$$A=\frac{ikn_0\mu(E_n-E_p)}{[Dk^2+(1/\tau_r)]^2+[k(Ec/H)-\omega]^2}\times\left[\left(Dk^2+\frac{1}{\tau_r}\right)-i\left(k\frac{Ec}{H}-\omega\right)\right]. \quad (7)$$

The corresponding space-charge field is

$$E_s=\frac{(D_p-D_n)dn/dy-n(\mu_nE_n+\mu_pE_p)}{n(\mu_n+\mu_p)}. \quad (8)$$

The deviation from charge neutrality required to set up this field is

$$(n-p)=\frac{\kappa}{4\pi e}\frac{dE_s}{dy}, \quad (9)$$

where κ is the dielectric constant. It is readily verified that the deviation from charge neutrality implied by Eq. (9) is normally small compared to $(n-n_0)$, as given by Eq. (7). For example, for $D_n=D_p$, $\mu_n=\mu_p$, and $Ec/H=s=\omega/k$, we find

$$\frac{n-p}{n-n_0}\left(\frac{\omega_c}{\omega_p}\right)^2\tau_r\left[Dk^2+\frac{1}{\tau_r}\right],$$

where $\omega_p{}^2=4\pi n_0e^2/m\kappa$. For bismuth at low temperatures $H\sim 10^4$ gauss, $\omega_c\sim 10^{12}$ sec^{-1}, $\omega_p\sim 10^{13}$ sec^{-1}, $D\sim 0.5$ cm^2/sec, $\tau\sim 10^{-9}$ sec, $\tau_r\sim 10^{-9}$ sec, one obtains $(n-p)/(n-n_0)\sim 1$ for $k\sim 5\times 10^5$ cm^{-1}. The above method of treating space-charge effects is therefore satisfactory for frequencies somewhat lower than 10^4 Mc/sec. It is to be noted that the criterion for validity is not strongly dependent on the magnitude of the magnetic field since $D\omega_c{}^2$ is independent of H.

The average energy transfer per unit volume, $\langle dW/dt\rangle$ is now readily calculated from Eqs. (2), (3), and (7), using the relation

$$\frac{dW}{dt}=\tfrac{1}{2}\,\mathrm{Re}[j_n'E_n{}^*+j_p'E_p{}^*]=\tfrac{1}{4}\,\mathrm{Re}[(j_n'-j_p')(E_n{}^*-E_p{}^*)]. \quad (10)$$

In Eq. (10), the primes denote the ac parts of the corresponding currents. We find

$$\left\langle\frac{dW}{dt}\right\rangle=\frac{en_0\mu[\mathcal{E}_n-\mathcal{E}_p]^2\epsilon^2k^2}{2}\times\left[\frac{1/k\tau_r(Dk+1/k\tau_r)-s(Ec/H-s)}{(Dk+1/k\tau_r)^2+(Ec/H-s)^2}\right]. \quad (11)$$

The corresponding attenuation constant α for the acoustic wave is obtained from $\alpha=-(1/sW)\langle dW/dt\rangle$, where $W=\frac{1}{2}\rho s^2\epsilon^2$ (ρ= density) is the energy of the wave. Thus,

$$\alpha=\frac{en_0\mu(\mathcal{E}_n-\mathcal{E}_p)^2k^2}{\rho s^3}\times\left[\frac{1/k\tau_r(Dk+1/k\tau_r)-s(Ec/H-s)}{(Dk+1/k\tau_r)^2+(Ec/H-s)^2}\right]. \quad (12)$$

In the limit $\tau_r\rightarrow\infty$ (no recombination), this result is similar to Eq. (1) of Hutson *et al.*[1] Acoustic gain is in this case obtained when $Ec/H>s$, provided attenuation resulting from other sources may be ignored. However, for finite recombination times, Eq. (12) shows that the onset of acoustic gain is somewhat delayed. The condition for onset of gain is, in this case,

$$\frac{Ec}{H}-s\geq\frac{1}{s}\left[\frac{D}{\tau_r}+\left(\frac{1}{k\tau_r}\right)^2\right]. \quad (13)$$

DISCUSSION

The magnitude of the attenuation constant α is primarily determined by the factor outside the brackets in Eq. (12). As an example, we shall consider a transverse mode in bismuth with $s=10^5$ cm/sec and $k\sim 10^4$ cm^{-1}($\nu\simeq 100$ Mc/sec), and $H=10^4$ gauss. The deformation potential constants $\mathcal{E}_n$, $\mathcal{E}_p$ are not known, however, as a reasonable value we shall take $\mathcal{E}_n-\mathcal{E}_p=10$ v for purposes of estimating. Using $\mu=50$ cm^2/v sec, we find

$$\alpha_0=en_0\mu(\mathcal{E}_n-\mathcal{E}_p)^2k^2/\rho s^3\simeq 30\text{ cm}^{-1}.$$

Moreover, the minimum value of the bracketed quantity is in a typical case ~ -2.5 (see Fig. 1) so that the maximum negative value of α is ~ -75 cm^{-1}.

This acoustic gain is large enough to overcome losses due to other effects, so that net acoustic gain of the order of 300 db/cm is attainable.

The attenuation constant α is plotted for $k=10^4$ cm^{-1}, $s=10^5$ cm/sec, $D=1$ cm^2/sec, and $\tau_r\sim 10^{-8}$ sec in Fig. 1. This value of τ_r is not unreasonable for bismuth at low temperatures, although it is possible that τ_r is considerably longer. A larger value of τ_r would result in a higher peak than is shown in Fig. 1. It should be possible to obtain a value of the bulk recombination time τ_r from observing the shift of the onset of acoustic gain with frequency [see Eq. (13)]. If τ_r and D are known, the above experiment might also be used to study the dependence of the velocity of sound on sample orientation.

Note added in proof. Hopfield has recently treated this problem, [Phys. Rev. Letters **8**, 311 (1962)], considering only the effect of the deformation potentials on local recombination. He correctly assumes that the carrier distributions will relax to equilibrium values determined by the local strain. If we incorporate a strain dependent recombination process into our theory the following changes will occur. For $n-n_0\ll n_0$ it can

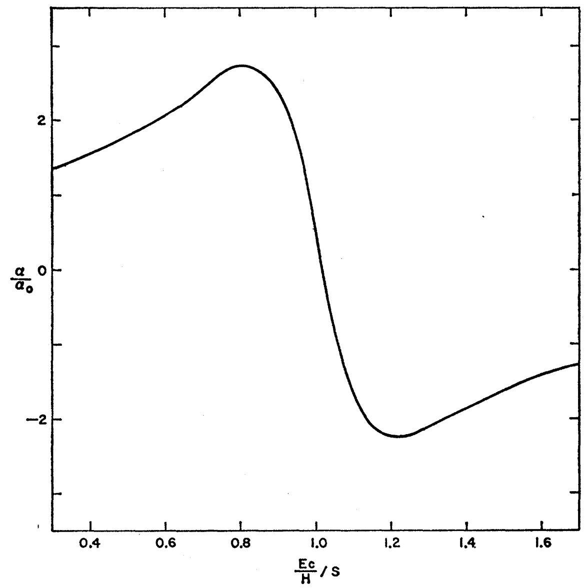

FIG. 1. Plot of bracketed term in Eq. (12) for the following values of parameters: $D=1$ cm^2/sec, $\tau_r=10^{-8}$ sec, $k=10^4$ cm^{-1}, $s=10^5$ cm/sec.

be shown that the recombination rate will be given by

$$dn/dt=(-1/\tau_r)[(n-n_0)+N_r\epsilon(\mathcal{E}_n-\mathcal{E}_p)],$$

where N_r is a reduced density of states given, in terms of the density of states at the Fermi level in the conduction and valence bands, by $N_r^{-1}=N_c^{-1}+N_v^{-1}$. If we use this recombination rate in Eqs. (5a) and (5b), the amplitude of $n-n_0$ is altered and in Eq. (7) the factor $n_0\mu$ is replaced by $(n_0\mu+N_r/k^2\tau_r)$. The form of the attenuation constant is not altered, but an additional term given by

$$\alpha'=\frac{eN_r(\mathcal{E}_n-\mathcal{E}_p)^2}{\tau_r\rho s^3}\times\left\{\frac{Dk[Dk+(1/k\tau_r)]-(Ec/H)[(Ec/H)-s]}{[Dk+(1/k\tau_r)]^2+[(Ec/H)-s]^2}\right\}$$

is now present and should be added to that given in Eq. (12). α' is similar to the expression given by Hopfield if diffusion is ignored. Because of the term $-Dk[Dk+(1/k\tau_r)]$, α' can be negative corresponding to amplification for Ec/H slightly less than s.

The ratio of the two contributions to the attenuation constant is approximately given by $\alpha/\alpha'\cong\tau_r n_0\mu k^2/N_r \cong\tau_r E_F\mu k^2$ where E_F is the average Fermi level in the valence and conduction bands. For sound waves with small k it is clear that the recombination which caused term α' will dominate the attenuation, whereas the transport term α will dominate at higher values of k. The value of τ_r which enters the ratio α/α' is presently unknown, although it is thought to be large ($\sim10^{-6}$ sec) since recombination employing phonons should be negligible at temperatures at which amplification should be observed and scattering by impurities between states far apart in momentum is highly improbable.

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Nonlinear Dielectric Polarization in Optical Media

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The physical mechanisms which can produce second-order dielectric polarization are discussed on the basis of a simple extension of the theory of dispersion in ionic crystals. Four distinct mechanisms are described, three of which are related to the anharmonicity, second-order moment, and Raman scattering of the lattice. These mechanisms are strongly frequency dependent, since they involve ionic motions with resonant frequencies lower than the light frequency. The other mechanism is related to electronic processes of higher frequency than the light, and, therefore, is essentially flat in the range of the frequencies of optical masers. Since this range lies an order of magnitude higher than the ionic resonances, the fourth mechanism may be the dominant one. On the other hand, a consideration of the linear electro-optic effect shows that the lattice is strongly involved in this effect, and, therefore, may be very much less linear than the electrons. It is shown that the question of the mechanism involved in the second harmonic generation of light from strong laser beams may be settled by experiments which test the symmetry of the effect. The electronic mechanism is subject to further symmetry requirements beyond those for piezoelectric coefficients. In many cases, this would greatly reduce the number of independent constants describing the effect. In particular, for quartz and KDP there would be a single constant.

THE recent observation by Franken, Hill, Peters, and Weinreich[1] of second-harmonic light generation in quartz raises the question of the physical mechanism involved in nonlinear dielectric polarization effects at optical frequencies. The purpose of this paper is to discuss briefly the possible types of mechanism, and to point out the special properties of one mechanism which may permit it to be experimentally separated

[1] P. Franken, A. Hill, C. Peters, G. Weinreich, Phys. Rev. Letters 7, 118 (1961).