

with a similar expression for  $T_R^{-1}$ . The frequency (22) may then be written as

$$\nu = \omega - (\nu/2Q)^{-1}(\omega - \Omega)(I/2e)(N_L^{-1} - N_R^{-1}), \quad (23)$$

which is the result (18).

Finally, let us estimate the magnitude of the frequency pulling,

$$\nu - \omega = (I/2e)[(N_L - N_R)/N_L N_R](\omega - \Omega)(\nu/2Q)^{-1}.$$

For a current of  $10^{-2}$  A,  $I/2e$  is of the order of  $10^{16}$ - $10^{17}$ . The difference in the number of pairs  $N_L - N_R$  may be estimated by knowing the capacitance of the junction ( $C \approx 1 \mu\text{F}$ ) and the voltage  $V$  ( $\approx 10^{-5}$  V), to be of the order of  $10^8$  pairs. The magnitude of  $N_L$  or  $N_R$  for a volume  $10^{-6}$  cm<sup>3</sup> is of the order of  $10^{12}$ . From these numbers we find

$$\nu - \omega \approx (1-10) \times (\omega - \Omega)(\nu/2Q)^{-1};$$

so that if  $(\omega - \Omega)$  is around  $\nu/2Q$ , the pulling would be between 1 and 10 Hz. As Eq. (18) seems to be very sensitive to the number of pairs on the right- and left-hand sides, it would be interesting to measure the frequency pulling for superconductive films of different thicknesses.

We would like to thank Professor L. W. Gruenberg and Professor P. C. Martin for helpful discussions, and Professor M. J. Stephen for collaboration on certain portions of this work.

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<sup>2</sup>B. D. Josephson, Phys. Letters **1**, 251 (1962).

<sup>3</sup>W. E. Lamb, Jr., Phys. Rev. **134**, A1429 (1964).

<sup>4</sup>M. O. Scully and W. E. Lamb, Jr., Phys. Rev. **159**, 208 (1967). References to other work will be found in the introduction of this paper.

<sup>5</sup>Several other models and calculational techniques have been used and lead to the same result. These will be reported in a more detailed publication.

<sup>6</sup>P. W. Anderson, Phys. Rev. **112**, 1900 (1958). In the present paper  $s^p$  corresponds to  $c - p^c p$ , where  $c_p$  is the single-particle annihilation operator.

<sup>7</sup>The voltage is related to the number of excess pairs. This however is not a fixed number [See Eq. (10)], and one expects fluctuations in the voltage. M. J. Stephen has shown (private communication and to be published) that these voltage fluctuations lead to a linewidth in agreement with the recent experiments reported by W. H. Parker [in *Fluctuations in Superconductors*, edited by W. S. Goree and F. Chilton (Stanford Research Institute, Stanford, California, 1968)].

<sup>8</sup>The effects of a finite cavity  $Q$  are discussed in Ref. 4. There we were working in the number representation; however, it is clear that Eq. (6) of the present paper follows directly from Eq. (84) of Ref. 4.

<sup>9</sup>Equation (7) was derived by considering the wires connecting the superconductors as particle reservoirs of unpaired electrons. The coefficient  $A$  may be related to the dc current by noting that  $I$  equals  $2e \langle \dot{S}_{Lz} \rangle$ . The detailed development of Eq. (2) will be given in future publication.

<sup>10</sup>Calculations of this type are to be found in M. Scully, thesis, Yale University, 1966 (unpublished), Appendix V.

## VARIATION OF CHARGE-CARRIER RELAXATION TIME IN BISMUTH WITH MAGNETIC FIELD\*

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The attenuation of Alfvén waves in bismuth can be analyzed to show the detailed variation of the relaxation time with magnetic field. For the example presented, which has the magnetic field along the threefold axis, variations between  $2.5 \times 10^{-10}$  and  $6.0 \times 10^{-10}$  sec are found around an average value for  $\tau$  of  $4.3 \times 10^{-10}$  sec.

The transmission of Alfvén waves in bismuth has been used in several laboratories to determine the effective masses and number of charge carriers.<sup>1,2</sup> In addition, the attenuation coefficient of Alfvén waves is directly related to the carrier relaxation times  $\tau$  (which are, in general, tensor quantities). In this Letter we show that in addition to average values for  $\tau$ , we can obtain good numerical values showing the variation in  $\tau$  with magnetic field using data on the at-

tenuation of Alfvén waves. This variation in  $\tau$  with magnetic field is, of course, responsible for the de Haas-Shubnikov effect.

Numerical values of  $\tau$  in bismuth from Alfvén-wave experiments have been obtained by McLachlan<sup>2</sup> and Marsten and Kao.<sup>3</sup> A theoretical study has been performed by Brownell and Hygh.<sup>4</sup> In each of these studies it is clear that the relaxation times are not isotropic. In each of the previous studies only the average value of the relax-

ation time for each orientation has been obtained. We have been able to obtain numerical values for the variation in relaxation time by using a thick (8.6-mm) sample, so that many interference peaks are present, and by analyzing the data in a somewhat different way than previous workers.

Figure 1 shows the experimental data analyzed in this Letter. The oscillations are caused by interference between microwaves which have passed once through the sample, and microwaves which have "leaked" around the sample. The Fabry-Perot type of pattern, discussed below, is not seen because of the thickness of the sample. The feature of the data under discussion in this Letter is the envelope of the interference pattern. This envelope has oscillations due to quantum oscillations in the relaxation time and thus in the Alfvén-wave attenuation. The period of these oscillations is the same as the period of the de Haas-Shubnikov oscillations.

As discussed in Refs. 1 and 2 the transmission shows a series of intensity maxima and minima. The exact number of these maxima depends on the interference mechanism which can be either of the Fabry-Perot or leakage type. We will develop an expression for the transmission coefficient including both leakage and Fabry-Perot-type interference.

Consider an electromagnetic wave incident on a disk of thickness  $d$ . The index of refraction of this disk is, neglecting terms of order  $1/\omega^2\tau^2$ ,<sup>1,2</sup>

$$N = (c/V)(1 + i/2\omega\tau), \quad (1)$$

where  $V$  is given by

$$V = H/[4\pi m_0 n f(m^*)]^{1/2}. \quad (2)$$

Here  $\omega$  is the experimental angular frequency,  $c$  the speed of light,  $V$  the Alfvén velocity,  $H$  the magnetic field,  $m_0$  the free electron mass, and  $n f(m^*)$  the Alfvén mass density.<sup>1,2</sup> The experimental conditions are such that  $c/V \gg 1$ ,  $\omega\tau \gg 1$ , and  $\omega_c > \omega$ , where  $\omega_c$  is the cyclotron frequency. This means that  $N$  is nearly real and  $|N| \gg 1$ .

Under the conditions of this experiment  $\omega\tau$  was greater than 25 and therefore the propagation velocity was not affected by the imaginary part of  $N$ . The imaginary part does govern the attenuation of the Alfvén wave propagated in the sample. In terms of Eq. (1) Alfvén waves in bismuth are attenuated in a sample with thickness  $d$  by the factor  $\exp(-d/2V\tau)$ .

In general the interference fringes are due to mixing of microwave power transmitted through the bismuth disk and microwave power that has "leaked" around the sample. The transmitted power is determined by multiple internal reflections in the bismuth disk. We are considering a microwave signal with an electric field of amplitude  $E_i$  incident on the bismuth slab. An amplitude  $E_r$  is reflected and that part which is not reflected divides into two parts, a fraction  $A$  which penetrates the sample, and a fraction  $B$  which leaks around the sample. The following expression for the transmitted wave may be found in standard optics textbooks<sup>5</sup>:

$$E_T = t_{12} t_{21} (1 + r_{21}^2 e^{i2\delta} + r_{21}^4 e^{i4\delta} + \dots) A E_i e^{i\delta}. \quad (3)$$

Here  $t_{12}$ ,  $t_{21}$ , and  $r_{21}$  are the transmission and reflection coefficients of the surface of the disk and  $\delta$  the phase change of the wave in traversing the disk.  $\delta$  is complex to allow for attenuation of the wave in the sample. These quantities are defined below:

$$r_{12} = -r_{21} \frac{N-1}{N+1}, \quad t_{12} = \frac{2}{N+1}, \quad t_{21} = N t_{12};$$

$$\delta = \frac{\omega d}{V} \left(1 + \frac{i}{2\omega\tau}\right) = \frac{\omega}{c} N d. \quad (4)$$

Because of attenuation in the sample each internal reflection is progressively attenuated. For bismuth of millimeter thickness in magnetic fields of 25 kG and less, this attenuation is sufficiently strong that only the first two terms in Eq. (3) contribute. The leakage wave is given by

$$E_L = B E_i e^{i\delta'}, \quad (5)$$

where  $B$  is the fraction of the incident wave that leaks around the sample and

$$\delta' = \omega d/c.$$

We have assumed, for convenience, that the phase velocity of the leakage wave is  $c$ . The total wave received at the detector is then

$$E_0 = [B e^{i\delta'} + t_{12} t_{21} A e^{i\delta} (1 + r_{21}^2 e^{i2\delta})] E_i, \quad (6)$$

and the transmission coefficient is defined as

$$T = \left| \frac{E_0}{E_i} \right|^2 = B^2 + 8AB \frac{V}{c} \exp\left(\frac{-d}{2V\tau}\right) \cos \omega d \left(\frac{1}{V} - \frac{1}{c}\right) + 16A^2 \frac{V^2}{c^2} \exp\left(\frac{-d}{V\tau}\right) \left[1 + 4\frac{V}{c} + 2 \exp\left(\frac{-d}{V\tau}\right) \cos \frac{2\omega d}{V}\right]. \quad (7)$$

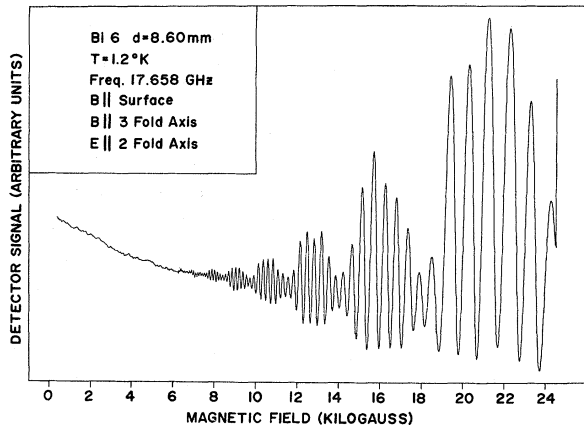


FIG. 1. Interference fringes in a sample of bismuth. The oscillations in the envelope reflect the oscillations in relaxation time discussed in the text.

The first two terms in Eq. (7) describe leakage transmission, and the third term describes Fabry-Perot-type transmission. The leakage term is an order of magnitude larger than the Fabry-Perot term. Since Fabry-Perot interference is due to multiple reflection, the second transmitted wave is very much weaker than the first, and only at large magnetic fields is it strong enough to cause observable interference. The leakage case requires only the first transmitted term to cause interference with the leakage wave; so interference occurs at lower magnetic fields. The present experiments were done with a sample 8.6-mm thick. This means that the multiply reflected waves should have negligible contribution so that no Fabry-Perot interference should be present. This was experimentally verified. Under these conditions the transmission coefficient is approximately

$$T \approx B^2 + 8AB \frac{V}{c} e^{-d/2V\tau} \cos \frac{\omega d}{V}. \quad (8)$$

The maxima and minima of  $T$  occur very nearly at  $\cos(\omega d/V) = \pm 1$ . This corresponds to changes in the "optical" thickness of the sample of one-half wavelength from maximum to minimum in the transmission. The transmission envelope is given by

$$T_{\max} - T_{\min} = \Delta T \approx (8ABd/\lambda_0 n) \times \exp(-n\pi/\omega\tau), \quad (9)$$

where  $\lambda_0$  is the free-space wavelength of the microwaves and  $n$  is the index of the transmission maxima. This expression does not contain the

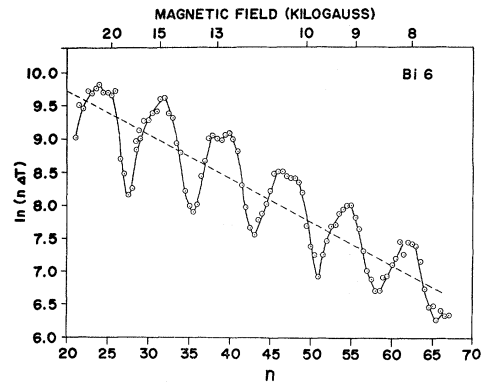


FIG. 2. A plot of  $\ln(n\Delta T)$  vs  $n$  for the data of Fig. 1. The dashed line is a least-squares fit to the data points.

Alfvén mass density; so quantum oscillations of the mass density do not affect the transmission envelope. A plot of  $\ln(n\Delta T)$  vs  $n$  should yield a straight line whose slope is an average  $\tau$  and whose intercept gives  $\ln(8ABd/\lambda_0)$ . When the experimental data are plotted in this manner, oscillations are found about the straight line. These have been mentioned before by Marsten and Kao<sup>3</sup> and McLachlan<sup>2</sup> and are due to oscillations in  $\tau$  with magnetic field. Figure 2 is a plot of the logarithm of  $n\Delta T$  vs  $n$  using Eq. (9). This figure shows clearly the period of the oscillations in relaxation time. This period is  $(1.56 \pm 0.05) \times 10^{-5} \text{ G}^{-1}$ , which agrees well with hole period for the trigonal direction measured using other oscillatory effects. We have studied other orientations but the oscillations in  $\tau$  are not as pronounced as for this case. These will be reported at a later date. The straight line in Fig. 2 is a least-squares fit to the experimental points. The slope of the line gives an average relaxation time of  $0.426 \times 10^{-9}$  sec. This value is in agreement with the experimental values found by McLachlan,<sup>2</sup> and Marsten and Kao<sup>3</sup> and with the theoretical values of Brownell and Hygh.<sup>4</sup> Obviously the use of a simple least-squares fit in this case is open to question, but it gives a preliminary value for  $\tau$  which can be refined.

To obtain values for the variation of the relaxation time with magnetic field the following procedure is followed. Manipulation of Eq. (9) yields the following expression for  $\tau$ :

$$\tau = \frac{\pi n}{\omega} \left( \ln \frac{8ABd}{\lambda_0 n \Delta T} \right)^{-1}. \quad (10)$$

The intercept of the plot in Fig. 2 yields  $\ln(8ABd/\lambda_0)$ , for use in Eq. (10). All other quantities in Eq. (10) are known. The numerical value of the

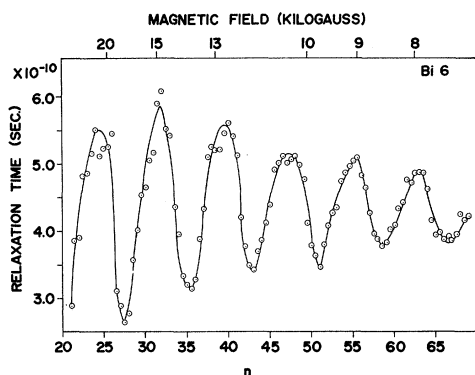


FIG. 3. A plot of relaxation time  $\tau$  vs  $n$  which is equivalent to  $\tau$  vs  $1/H$ .

intercept in Fig. 2 is  $11.114 \pm 0.003$ . This has been used to find values for  $\tau$  as a function of magnetic field. These results are plotted in Fig. 3, which gives  $\tau$  as a function of  $n$ , which is equivalent to  $\tau$  as a function of  $1/H$ .

In Fig. 3 we see variations in  $\tau$  between  $2.5$  and  $6.0 \times 10^{-10}$  sec around the average value of  $4.26 \times 10^{-10}$  sec. The absolute accuracy of these numbers is believed to be within about 10%. The precision with which this technique shows the variations in  $\tau$  is, according to Fig. 3, much bet-

ter than 10%. The primary contribution of this Letter lies in demonstrating numerically the dramatically large variation in  $\tau$  with magnetic field. Since the period observed is the same as the period of the holes observed in de Haas-Shubnikov and de Haas-van Alphen experiments, the variations shown here are presumed to be variations in the hole relaxation time.

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## ROOM-TEMPERATURE ELECTROPHOTOLUMINESCENCE IN KCl

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Some data showing the existence of electrophotoluminescence in alkali halides at room temperature are reported. The effect is studied for  $h\nu > E_g$  in KCl(KI) and a model which relates it to variations of the absorption coefficient is proposed.

In this Letter we wish to give some experimental evidence of the existence of electrophotoluminescence in alkali halides at room temperature and show that this effect may be useful in studying the nature of the strong absorption peaks above the onset of the interband transitions.

It is well known from the literature<sup>1,2</sup> that alkali-halide crystals luminesce in the visible and near-ultraviolet when irradiated with photons with  $h\nu > E_g$ . The room-temperature photoluminescence produced in this way was explained in terms of recombination between optically created free holes and electrons trapped at  $F$  centers formed by means of ultraviolet coloration.<sup>3</sup> The crystals we used were Harshaw KCl strongly doped with KI. In these crystals the parameters

related to the equilibrium concentration of the  $F$  centers have been determined by Goldberg and Mahr.<sup>4</sup> The photoluminescence spectra are perfectly similar to those reported in Ref. 3.

A freshly cleaved surface of the crystal, 1.5 mm wide and 8 mm thick, was irradiated with light from a vacuum monochromator having a resolution of 25 Å. A dc source was employed so that the radiation was constant in time. The ac field was applied parallel to the surface using plane capacitor plates covered with a very thin insulating layer in order to prevent carrier injection.<sup>5</sup> A RCA 1P28 phototube was put close to the sample on the path of the emitted and reflected light, which were separated by means of filters. Phase-sensitive detection was used. The crys-