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Non-Ohmic Conduction in Bismuth

Abstract: The experimentally determined I-V characteristics at large currents at 4.2°, 77°, and 300°K have been found to be nonlinear in both single crystal and wire samples of bismuth. A theory has been formulated which includes self-magnetoresistance, a redistribution of carriers (pinch) and bimolecular bulk and surface recombination. Very little information exists, either experimentally or theoretically, with regard to these recombination mechanisms. A program has been written to numerically solve the equations for various values of the electron-hole recombination time and surface velocity. It is found that the I-V characteristics are dominated by the self-magnetoresistance and vary only slightly for a wide range of the generation-recombination parameters.

Introduction

The experimental investigation reported in this paper was motivated by an interest in the interband recombination of electrons and holes in a semimetal such as bismuth under conditions which require a large change in crystal momentum with essentially no change in energy. Interband transitions may be induced by impurities or defects, absorption or emission of a phonon, or absorption of a photon and emission of a phonon, or be multi-particle transitions. Since the energy of a phonon of the required wave vector is about 50°K,[1] the only significantly frequent transitions in a crystal at liquid helium temperatures (and sufficiently shielded from 77°K radiation) can be those induced by impurities, defects, and possibly the simultaneous transition of two Coulomb-interacting electrons.

The above suggests that the recombination time at liquid helium temperatures in pure and perfect bismuth crystals should be quite long—long enough so that it could be measured by a pinch-effect experiment. If the electron-hole plasma is pinched in a time short compared to the recombination time, the subsequent current should show the reappearance of carriers in the region of the crystal from which they had been swept. It was hoped that this time dependence could be observed in the variation of the current produced by the application of an appropriate voltage step.

The large current required for pinching determines that fast voltage pulses be used. However, the usual difficulties, sample inductance, skin effect and the generation of sufficiently large rectangular pulses, combine to make a direct measurement of the generation time very difficult. More significantly, because of magnetoresistance, the large spatially dependent self-magnetic field produced by the current causes a non-ohmic variation of current with voltage, which retards the pinching. The detailed variation of the current with voltage is, however, a function of the bulk and surface recombination times. Reported here is an attempt at determining these recombination times by comparing experimental I–V data with numerical solutions of the steady state conductivity for a range of values of the bulk and surface recombination times. For convenience these experiments were started at 77° and 300°K; some 4° data was also taken. It turns out that the I–V characteristics are dominated by the self-magnetoresistance, and vary but slightly for a wide range of the generation-recombination parameters, even though the computed results show large changes in the radial distribution and total number of carriers.

Experimental details

The experimental apparatus consisted of a pulse generator connected to a delay line terminated with the sample. The pulses were generated by discharging a 10-ohm pulse forming line (five 50-ohm coaxial cables in parallel) through a mercury relay. The line could be charged to 5 kilovolts, thereby producing a current pulse of 250 amperes. The sample appears as a short circuit across the

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delay line and causes an almost equal and opposite reflected pulse which doubles the current through the sample. A Tektronix 541 oscilloscope was connected to the larger end contacts of the sample to measure the sample voltage. The current is, of course, determined from the line impedance and charging voltage.

The experimental data were obtained by varying the charging voltage of the pulse forming line in steps and measuring the sample voltage. Typical data at 77° and 300°K for a (polycrystalline) Bi wire sample are shown in Fig. 1. The next higher 300°K data point taken destroyed the sample, probably by the mechanism of thermal pinching and melting recently described by Ancker-Johnson and Drummond.[2] Similar I–V data have been obtained on single crystal samples at 4.2°, 77° and 300°K, but it has not been possible, to date, to get data on a sample whose axis is in the trigonal direction. For this direction, the theory is particularly simple but the samples are very fragile.

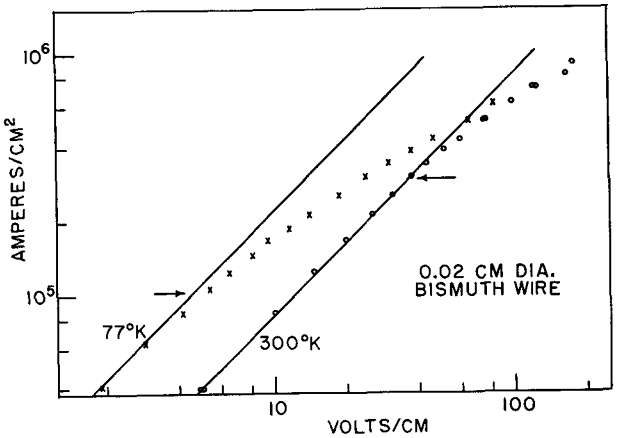


Figure 1 **Experimental data taken on the same (polycrystalline) bismuth wire sample at 77° and 300°K.** *The straight lines are an extrapolation of the ohmic range; the arrows indicate the classical pinching current* $I_p = \sqrt{4\pi n_o \xi a^2}$.

Theory

We consider a steady state pinch in a cylindrical sample with its axis along the trigonal direction. Both the linear drift and Hall conductivities in the radial direction (the trigonal plane) are isotropic; this greatly simplifies the computational problem. It is assumed the radius a is large compared to the shielding length and the mean free path. A current flowing along the axis of the sample will produce a circumferential magnetic field[3] H, within the sample,

$$H(r) = \frac{4\pi e E}{r} \times \int_0^r \left[\frac{\mu_3}{1 + a_1^2 H(r')^2} + \frac{\nu_3}{1 + b_1^2 H(r')^2}\right] n(r') r' \, dr', \quad (1)$$

where $n(r')$ is the carrier concentration, E is the applied electric field; $a_1^2 = \frac{1}{2}(\mu_1 + \mu_2)\mu_3 - \mu_4^2$ and $b_1^2 = \nu_1 \nu_3$ are the Hall mobilities in terms of the partial mobilities of one ellipsoid (μ_1, μ_2, μ_3, μ_4 for electrons and $\nu_1 = \nu_2$, ν_3 for holes). This magnetic field gives rise to Lorentz forces on the moving carriers which push both holes and electrons to the axis. The Lorentz forces are balanced by both diffusion and a Hall field E_H. The drift velocity for holes and electrons in the radial direction is then

$$v_p = -D_p(H)\frac{d \ln n}{dr} - \frac{\nu_1 \nu_3 E H}{(1 + b_1^2 H^2)^2} - \frac{\nu_1 E_H}{1 + b_1^2 H^2},$$

$$v_n = -D_n(H)\frac{d \ln n}{dr} - \frac{(\mu_1 + \mu_2)\mu_3 E H}{2(1 + a_1^2 H^2)^2} + \frac{(\mu_1 + \mu_2) E_H}{2(1 + a_1^2 H^2)}. \quad (2)$$

In the steady state, the radial velocities and the density distributions of the holes and electrons must be the same, since the space charge forces are sufficiently strong to maintain charge neutrality within a shielding length. Equating the velocities determines the Hall field, which may then be eliminated. The Einstein relation appropriate for Fermi statistics, $D = 2\mu\xi/3e$, where ξ is the Fermi energy, is used to specify the magnetic field dependence of the diffusion constants.

The steady-state radial particle flux is then

$$N = \frac{(\mu_1 + \mu_2)\nu_1}{(\mu_1 + \mu_2 + 2\nu_1)(1 + \mu^2 H^2)e} \times \left\{2\xi \frac{dn}{dr} + \frac{ne(\mu_3 + \nu_3)E(1 + c^2 H^2)}{(1 + a_1^2 H^2)(1 + b_1^2 H^2)}\right\}, \quad (3)$$

where

$$\xi = [2\mu_3 \nu_1 \xi_n + (\mu_1 + \mu_2)\nu_3 \xi_p]/3(\mu_1 + \mu_2)\nu_3,$$

$$\mu = [(\mu_1 + \mu_2) b_1^2 + 2a_1^2 \nu_1]/(\mu_1 + u_2 + 2\nu_1),$$

$$c^2 = [\nu_1 \nu_3 (\mu_1 + \mu_2) + 2a_1^2 \nu_1]/(\mu_1 + \mu_2 + 2\nu_1)$$

and ξ_n, ξ_p are the electron and hole Fermi energies. Assuming a bimolecular energy independent generation-recombination term, we have a steady-state continuity equation

$$\frac{dN}{dr} + \frac{N}{r} + \frac{n^2 - n_0^2}{2\tau} = 0. \quad (4)$$

Substituting (3) into (4) yields, after eliminating dH/dr by means of the derivative of (1),

$$\frac{d^2 n}{dr^2} + \frac{dn}{dr}\left[\frac{1}{r}\left(\frac{1 + 3\mu^2 H^2}{1 + \mu^2 H^2}\right) + \frac{4EH\mathcal{H}_1\mathcal{H}_2}{aE_p H_p}\right.$$

$$+\left(1-\frac{n\mu^3 H_p^2 H}{n_0(1+\mu^2 H^2)}\right)\Bigg]+\frac{16n^2E^2\mathcal{H}_1^2}{a^2E_p^2n_0}(\tfrac{1}{2}-\mathcal{H}_2)$$

$$+\frac{8nEH\mathcal{H}_1\mathcal{H}_2}{raE_pH_p}=\frac{(n^2-n_0^2)(1+\mu^2H^2)}{2n_0L^2} \tag{5a}$$

and

$$\frac{dH}{dr}=\frac{2EH_p\mathcal{H}_1 n}{aE_pn_0}-\frac{H}{r}, \tag{5b}$$

where

$$\mathcal{H}_1=(1+c^2H^2)/(1+a_1^2H^2)(1+b_1^2H^2)$$

$$\mathcal{H}_2=\frac{\mu^2H^2}{1+\mu^2H^2}+\frac{a_1^2H^2}{1+a_1^2H^2}+\frac{b_1^2H^2}{1+b_1^2H^2}-\frac{c^2H^2}{1+c^2H^2}.$$

H_p is defined as the magnetic field for which the energy density $H_p^2/8\pi$ equals an average carrier energy density $2n_0\xi$, L is an average diffusion length given by

$$L^2=2\tau[2\mu_3\nu_1\xi_n+\nu_3(\mu_1+\mu_2)\xi_p]/e(\mu_1+\mu_2+2\nu_1),$$

and E_p, the pinching field if the sample remained Ohmic, equals $a\sqrt{4\pi\xi n_0/2n_0(\mu_3+\nu_3)}$.

The two equations (5) must be solved simultaneously subject to the physical boundary conditions of zero magnetic field and zero derivative of the carrier concentration on the axis. The third boundary condition, the carrier concentration on the axis, is determined by the condition that the total particle flux at the surface be zero. This is accomplished by equating the particle flux at the surface to a bimolecular surface recombination term:

$$N(a)=S[n(a)^2-n_0^2]/2n_0.$$

There is some experimental evidence relating to the value of S. The accepted explanation of Esaki's "kink effect"[4] can only hold if $S \gtrsim 1\times10^5$ cm/sec, the sound velocity. An upper limit for S at 4°K for a smooth surface is set by the observation that surface scattering is mostly specular;[5] this limit for a smooth surface is an order of magnitude greater than the sound velocity. At higher temperatures, S is presumably greater still.

In the actual computations, a value for the carrier concentrations on the axis is assumed, and the equations integrated out to the surface. The particle flux and surface recombination term are then compared and the process iterated. The calculations were done numerically on an IBM 1620 computer, using the Hamming method. Figure 2 shows the computed I–V characteristics for a bismuth sample at 77°K for some limiting values of the surface and bulk parameters. The center curve, $L = 0$, is the ordinary self-magnetoresistance effect with no carrier redistribution. The upper and lower curves, $L = a$, correspond to a surface recombination velocity of infinity and the sound velocity, respectively, and a diffusion length L equal to the radius. For a 0.01 cm radius sample this corresponds to the rather large value of 6×10^{-8} sec for the bulk recombination time.[6] If the recombination time is reduced by a factor of ten (6×10^{-9} sec) the I–V curves are just barely distinguishable from the $L = 0$ curve, making it impossible to obtain a unique value for the recombination parameters by curve fitting to the experimental data.

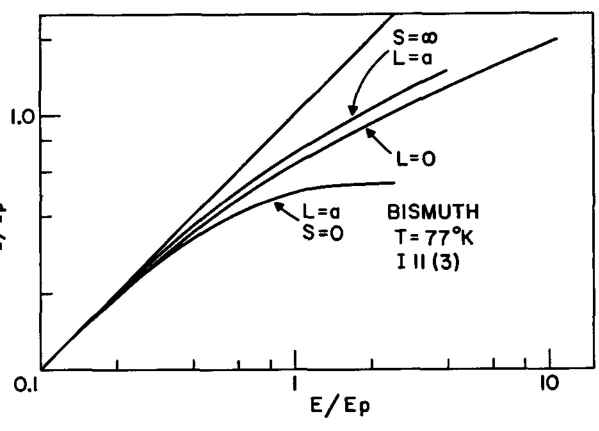
As the temperature is lowered, the mobility increases causing the self-magnetoresistance to become important at lower currents. The pinching current however remains unchanged and the deviations caused by the finite recombination time will be lost in the much larger self magnetoresistance effect. At higher temperatures, the pinching current increases owing to an increase in the number of carriers and a change from Fermi to Boltzmann statistics. This effect offsets the dropping mobility and again makes the deviations caused by a finite recombination time impossible to separate from the ordinary self-magnetoresistance.

In conclusion, the author wishes to thank S. H. Koenig for suggesting this investigation, for many informative discussions and for comments on the manuscript.

References

1. J. L. Yarnell, J. L. Warren, R. G. Wenzel, and S. H. Koenig, *IBM Journal* 8, 234 (1964). (This conference).
2. B. Ancker-Johnson and J. Drummond, *Phys. Rev.* 131, 1961 (1963).

Figure 2 **Computed steady-state I-V characteristics for some limiting values of the recombination parameters.** *For a 0.01 cm radius sample, L = a corresponds to a recombination time of 6 × 10⁻⁸ second.*



3. In computing the magnetic field we have neglected the small terms due to a non-zero tilt angle, which give rise to a transverse voltage component even in H. These terms go to zero in both the high and low field limits and presumably are not too important at intermediate fields.
4. L. Esaki, *Phys. Rev. Letters* **8**, 4 (1962).
5. A. N. Friedman and S. H. Koenig, *IBM Journal* **4**, 158 (1960).
6. A similar transition in Ge, where the relative parameters are not vastly different, takes 10^{-10} to 10^{-9} second. G. Weinreich, T. M. Sanders, Jr., and H. G. White, *Phys. Rev.* **114**, 33 (1959).

Discussion

M. Lampert: How can you directly detect self-pinching in the sample?

W. Schillinger: By measuring the spreading resistance of a point in contact with the side of the sample. If the carriers pinched, the spreading resistance would increase. However, to observe this effect in our samples would have required the surface conductivity to drop to 1% of the bulk value. No effect this large was seen.

W. R. Datars: We have observed non-Ohmic effects due to Joule heating in arsenic, antimony and bismuth. Do you have any assurance that such effects are not interfering with your observations?

Schillinger: The voltage across the sample (after an initial spike) had the same shape as the current through it, indicating that the conductivity during the pulse was constant and that heating effects were unimportant. This conclusion is substantiated by estimates of adiabatic heating during the pulse.