Electron Transport Properties of KBr in High Fields at Low Temperature*

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Transient photoconductivity measurements have been made on KBr crystals containing F centers as a function of electric and magnetic field at low temperature. The wavelength dependence of the observed photocurrent is discussed in terms of the known properties of F centers. Transverse-magnetoconductivity results were obtained which indicate that the conduction band of KBr is isotropic. Hot-electron effects were also observed at low temperature and were compared with an approximate theory involving a mixture of scattering mechanisms. Reasonable values of the low-field mobility parameters are obtained which can be used for interpretation of cyclotron-resonance experiments.

I. INTRODUCTION

THE cyclotron resonance of photoelectrons has recently been observed in crystals of KBr by an electron hearing technique.[1] These experiments were preceded by a detailed study of low-temperature transport processes in high electric and magnetic fields. Certain results of this study bear on the analysis of the cyclotron-resonance line shape as reported in the following article.[1] Other results, for example those on the angular dependence of transverse magnetoconductivity, relate to the shape of the conduction band and can be compared with band calculations of the type which have recently been carried out on KCl[2] and on KI.[3]

In the present experiments transient currents were produced in otherwise insulating crystals by optical excitation of F centers. Observations were carried out at different wavelengths corresponding to the F, K, and L bands, and it was found that ultraviolet excitation in the L-band region is relatively free of thermal or of field-ionization effects.[4] The photoelectrons are injected into the continuum at energies well above the bottom of the conduction band, but they rapidly cascade to the band minimum. F centers are the principal donors in these experiments, and since the ground state of the F center is more than 2 eV below the conduction band minimum, impact ionization is unimportant. The system is well suited to the study of hot-electron phenomena at moderate fields. It is however assumed that the trapping probability of a photoelectron (mainly at F centers) is relatively independent of field. This is a reasonable assumption for the low fields employed considering the trapping properties of F centers at low temperature.[5]

Non-Ohmic transient photoconductivity was first observed by Masumi[6] in AgCl crystals at low temperature. In the present work on KBr at 4.2°K, departures from Ohm's law were observed for electric fields above about 90 V/cm. Experiments were carried out to make sure that the observed phenomena was due to heating by the applied electric field and not to partial saturation or collection effects of the type observed by Lehfeldt[7] for the silver and thallium halides. The hot-electron phenomena in KBr are similar to those recently reported for KCl by Nakazawa and Kanzaki[8] and for CdS by Onuki and Shiga.[9]

II. APPARATUS

The transient photocurrents were detected using a method described by Van Heyningen[10] and applied to the magnetoconductivity of AgBr by Tippins.[11] The apparatus is shown schematically in Fig. 1. A cleaved and additively colored crystal of KBr (10^{16} F centers per cm^3) was sandwiched between closely spaced plane-parallel blocking electrodes. In some experiments the geometry was such that the KBr crystal could be illuminated from the side between the electrodes; in other experiments the light passed through the top transparent NESA electrode. The source of illumination was a quartz-prism monochromator equipped with either a quartz-iodine light source or an Osram HBO-200 discharge lamp. A photographic shutter was arranged to produce single pulses of light of several seconds duration and very low intensity. Photocurrents were detected by means of a Cary model 31 vibrating-reed electrometer used with the head of the instrument in the open position. The rate of drift with and without

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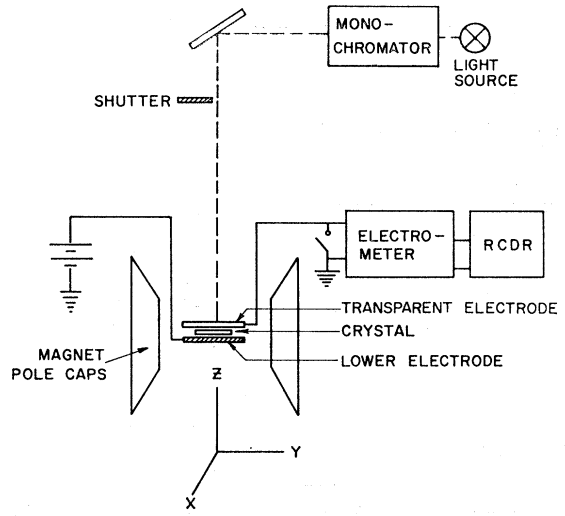

FIG. 1. Schematic diagram of the apparatus for the photocurrent measurements. The crystal is placed in a cryostat which is not shown. An electric field is produced by the battery and two parallel electrodes, one of which is semitransparent. A magnetic field perpendicular to the electric field is supplied by the magnet. The crystal is illuminated with pulses of light from the shutter, monochromator, and light source. The photocurrents are detected by a vibrating-reed electrometer.

exciting light was determined by measuring slopes on a recorder tracing, each corresponding to a light pulse of several seconds duration. In this way it was possible to work with very small amounts of charge and very low currents ($<10^{-14}$ A), thus avoiding polarization difficulties. Transverse magnetic fields up to 18 kOe were supplied by an electromagnet mounted on a rotating turntable. In some experiments magnetic fields up to 57 kOe were produced by means of a superconducting solenoid.

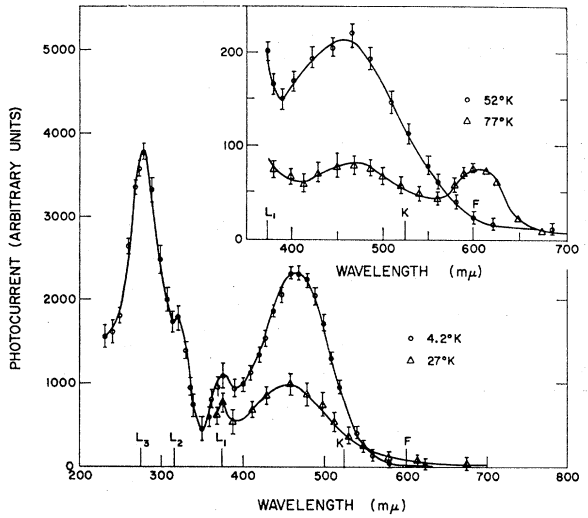

FIG. 2. The photoconductivity of a KBr sample containing 10^{16} F centers per cm^3 is shown versus the wavelength of the exciting light. Data for 4.2 and 27°K are shown in the main part of the figure, whereas the smaller currents for 52 and 77°K are shown in the insert. The positions of the optical absorption bands F, K, and L are also shown.

The sample itself was actually mounted within a small chamber sealed with Wood's metal and immersed in liquid helium. Heat transfer between the crystal and liquid helium was effected by having a small amount of helium exchange gas within the sample chamber. Temperatures were checked with a gold-cobalt: normal-silver thermocouple.

III. EXPERIMENTAL RESULTS

The transient photoconductivity of an additively colored crystal of KBr containing 10^{16} F centers per cm^3 is shown in Fig. 2. The photocurrent is shown in arbitrary units and no correction is made for absorption constant. Data for four different temperatures are shown and it can be seen that, except in the F band, the ultraviolet response increases with decreasing temperature due to increasing mobility as the lattice vibrations are frozen out.[4] The response peaks at 4.2°K can be identified with the previously reported positions of the L_1, L_2, and L_3 bands.[12] On the other hand, the peak at 460 mμ is in the high-energy tail of the so-called K band. The maximum in photoconductivity in this region is associated with a decreasing absorption constant but a rapid increase in quantum efficiency due to excitation to final states which overlie the continuum.[4] This peak can be identified at 77 as well as at 4.2°K and there seems to be little or no shift in wavelength with temperature. A similar peak was found at 470±10 mμ in RbCl at 4.2°K where the K and L_1 bands occur at 523 μ and 402 mμ, respectively.

Figure 3 shows the photoconductivity versus electric field for KBr at 77°K. The current is linear for excitation at 314 mμ but increases as $E^{1.3}$ for excitation at 500 mμ and for electric fields greater than about 1500 V/cm. At 500 mμ the quantum efficiency for exciting electrons into the conduction band is just starting to increase with photon energy.[4] This implies that some electrons are excited into states just below the conduction band. A high enough electric field should assist in the ionization of such electrons. A super-linear behavior is then to be expected. On the other hand, at $\lambda=314$ mμ the quantum efficiency is constant and high because the final states overlie the continuum at this wavelength. A linear dependence of current upon electric field is obtained. This is as expected at 77°K where the electron mobility is only about 200 cm^2/V sec.[13] Hot-electron effects are absent and apparently the electron-trapping probability is not strongly dependent upon external applied field.

Figure 4 shows the photocurrent as a function of electric field at 4.2°K with and without the presence of a transverse magnetic field. Three sets of data are

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shown corresponding to illumination at 500, 375, and 314 mμ. The critical field at which the $H=0$ currents start to deviate from Ohm's law occurs at about 90 V/cm for all three cases. However, the electric field dependence of the current at higher fields goes approximately as $E^{0.69}$ for $\lambda=500$ mμ, and as $E^{0.60}$ for $\lambda=375$ and $\lambda=314$ mμ. The difference in these two field dependences is thought to be due to field ionization of electrons from states just below the conduction band, as discussed in connection with Fig. 3. The field dependence at the shorter wavelengths is due primarily to a hot-electron effect.

The photocurrent versus electric field was also measured for crystals ranging in thickness from 0.44 to 2.66 cm. In all cases the critical field occurred at about 90 V/cm. This would rule out any partial saturation effect of the type observed by Lehfeldt[7] in AgBr. In that case, the distance that the electrons were able to travel before being trapped became comparable to the thickness of the crystal, i.e., all the electrons released by the light would be able to reach the anode and, thus, there would be no further increase in current with any increase in voltage.

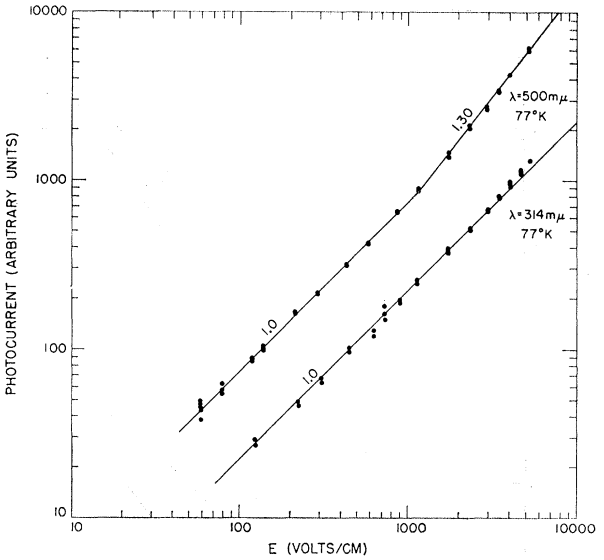

FIG. 3. The photocurrent versus the electric field is shown for the exciting light of wavelength 500 and 314 mμ at 77°K. The spectral bandwidths for the two wavelengths are 14 and 16 mμ, respectively.

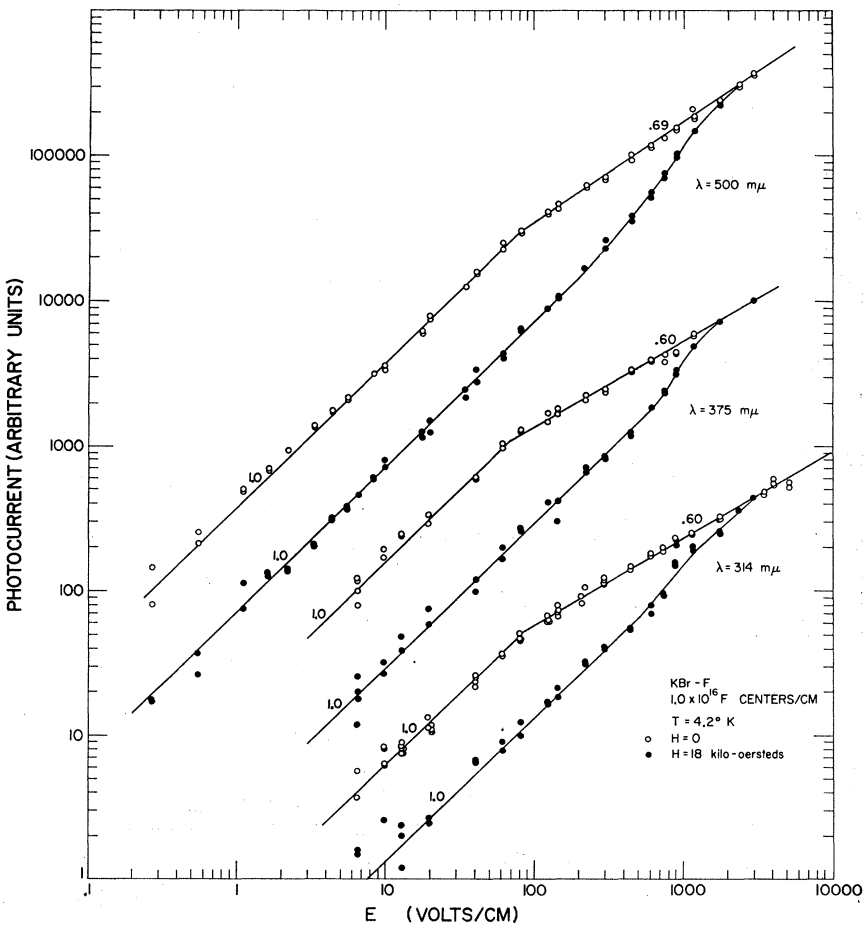

FIG. 4. The photocurrent versus the electric field is shown for KBr crystals containing 10^{16} F centers per cm³ at 4.2°K. The electric field dependence of the photocurrent $i(0)$ changes from $E^{0.69}$ at $\lambda=500$ mμ to $E^{0.60}$ at $\lambda=375$ and 314 mμ. The spectral bandwidths of the exciting light are 14, 16, and 16 mμ, respectively. The vertical placement of the pairs of curves for the three different wavelengths is arbitrary.

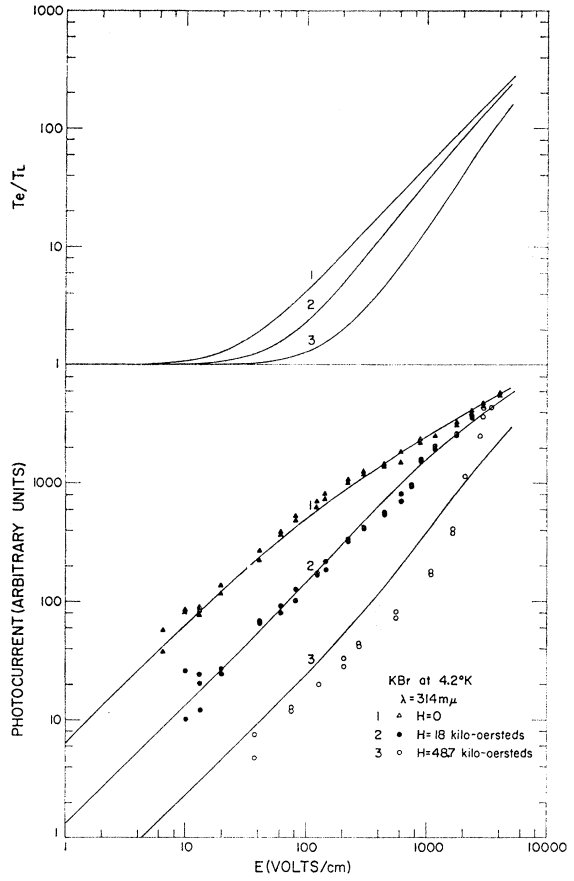

FIG. 5. Experimental points and curves from an energy-balance theory are shown for photocurrent versus electric field for different values of transverse magnetic field at 4.2°K. The theoretical fit is obtained using only acoustic and neutral impurity-scattering mechanisms. The parameters used are $\mu_0^{ac}=40\,000$ cm^2/V sec and $\mu_0^n=15\,200$ cm^2/V sec. The corresponding theoretical electron temperatures are shown on the top graph.

The data in the lower part of Fig. 5 were taken for transverse magnetic fields of $H=0$, 19, and 48.7 kOe. Curves computed from an approximate theory are also shown and these will be discussed in the next section.

Previous work has shown that a large fraction of the neutral defects or F centers in an alkali-halide crystal can be converted by F-band illumination at an intermediate temperature (120°K) to pairs of charged defects, F' and α centers. This has the effect of drastically reducing the low-temperature Hall mobility.[13] One can also see from curves of $i(0)/i(H)$ that the nature of the scattering mechanism can be altered. This is shown in Fig. 6 for a crystal at 4.2°K before and after conversion to F' and α centers. The definite rise in $i(0)/i(H)$ with increasing voltage at very-low electric field is indicative of a scattering mechanism which resembles charged impurity scattering. Initially, the crystal contained very few, if any, charged scattering centers. Such centers are not produced in great number by illumination at liquid-helium temperature.

Finally we show data on the effect of a transverse magnetic field as the field direction is changed with respect to the crystal axis. Such data bear on the isotropy of the conduction band and scattering mechanisms. In Fig. 7 we plot $i(H)/i(0)$ versus magnet angle θ for $H=8.0$ kOe in a plane parallel to one of the cube faces. An electric field of 112 V/cm was applied in the [001] direction. These results indicate that the conduction band of KBr is nearly isotropic and very probably of standard form.

IV. DISCUSSION OF RESULTS

Transient photocurrent measurements in the Ohmic region at low electric fields can be analyzed in a number of different ways. Perhaps the most appropriate is in terms of induced charge proportional to the product of the number of electrons released times the *Schubweg* defined as $w=\mu E\tau_t$, where μ is the mobility, E electric field, and τ_t the mean free time before trapping. From the work of Ref. 5 it appears reasonable to take τ_t as approximately constant. An equivalent approach is to consider the charge collected per unit time or current, and to carry out the analysis in terms of conductivity coefficients.[14] Assuming that a meaningful relaxation time $\tau(\epsilon)$ (where ϵ is electron energy) can be defined, it can be shown that the solution to the Boltzmann equation in the isotropic case yields the following expression for the current density[15]:

$$\mathbf{j}=\sigma(H)\mathbf{E}+\alpha(H)(\mathbf{E}\times\mathbf{H})+\gamma(H)(\mathbf{H}\cdot\mathbf{E})\mathbf{H}. \quad (1)$$

This equation is accurate to first order in $\mathbf{E}$ and to all orders in $\mathbf{H}$, a consequence of standard band shape and isotropic scattering. The coefficients σ, α, and γ are even functions of $\omega_c\tau$, where $\omega_c=eH/mc$ is the cyclotron frequency and τ is the relaxation time (the reciprocal of which can be written as a sum of different scattering probabilities).

In the present experiments the applied electric field $\mathbf{E}$ lies in the vertical or z direction (see Fig. 1) and the component of the current is measured in this same direction. A transverse Hall field is not allowed to build up because of the very-small transient current pulses involved. The transverse magnetic field $\mathbf{H}$ was applied in the x-y plane and was perpendicular to $\mathbf{E}$. Thus Eq. (1) gives the current in the z direction as

$$j_z=\sigma(H)E_z, \quad (2)$$

where

$$\sigma(H)=\frac{nq^2}{m}\left\langle\frac{\tau}{1+(\omega_c\tau)^2}\right\rangle. \quad (3)$$

In this last expression $q=-e$, the charge of an electron, m is an effective mass, and the brackets indicate a

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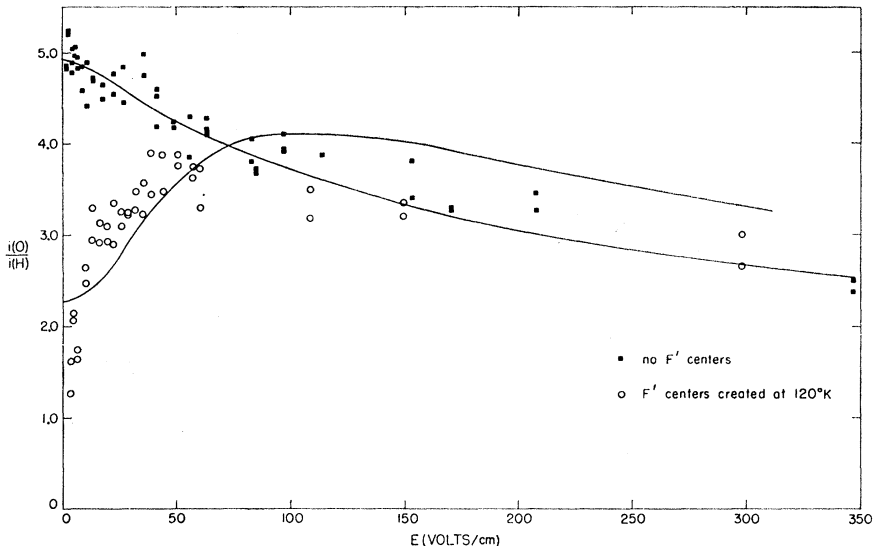

FIG. 6. The experimental points and curves from an energy-balance theory are shown for the ratio $i(0)/i(H)$ of the photocurrents with no magnetic field and with a magnetic field of 18 kOe as a function of electric field at 4.2°K. The dark squares show the values of $i(0)/i(H)$ for a crystal containing only F centers, and the corresponding theoretical curve was calculated using only acoustic and neutral impurity scattering mechanisms. The open circles represent the experimental values of $i(0)/i(H)$ for a crystal containing both F' and F centers. The F' centers were created by illuminating the crystal with F band light at 120°K. A typical theoretical curve using ionized impurity, neutral impurity, and acoustic-scattering mechanisms is also shown. Note that the experimental values rise much faster and drop to lower values at higher electric fields than the theoretical curve.

standard average over the carrier distribution. The quantity n is the average density of free carriers during a light pulse, a very small number as a consequence of a low generation rate and a very short trapping time. The light pulse duration was of the order of several seconds, during which time the integrated charge due to the drift of about 10^6 electrons was observed (in defining an electron distribution a time average rather than an ensemble average is involved).

Magnetoconductive effects proportional to the square of H become observable when the second term in the denominator of Eq. (3) becomes comparable to 1. If anisotropy in either effective mass or scattering probability is present it can be revealed by proper choice of crystal axes and rotation of the transverse magnetic field. Such effects were not observed although they can occur even in the case of a cubic crystal at high field (but not in the limit $H=0$).

It is also true that the magnetic field can conceivably affect the conductivity through a spin dependence of either scattering time[16] or trapping time. Such effects, if present, were small in the crystals studied since little or no *longitudinal* magnetoconductivity was observed.

Our remarks so far apply to the region of Ohmic conductivity at low electric field. Galvanomagnetic phenomena for hot electrons have been treated by a number of authors using the Boltzmann equation.[17] Customarily one retains only the first two terms of an expansion of the distribution function in Legendre polynomials of the angle θ between the momentum vector and the applied electric field. Considerable complication arises in the case of a mixture of scattering mechanisms. For example, elastic scattering combined with acoustic phonon scattering through deformation and piezoelectric interactions has recently been treated by Crandall.[18] Elastic and optical phonon scattering has been handled for semiconductors by a path variable method.[19] It is not yet clear whether these treatments can be extended to our case of an ionic crystal and especially to the cyclotron-resonance line-shape problem of the following article.[1]

There does exist a much more tractable but approximate method of analysis which is based upon the concept of effective electron temperature. This approach was introduced by Shockley,[20] who assumed that the steady-state electron distribution is Maxwellian with a mean energy $\frac{3}{2}kT_e$. The effective electron temperature T_e is obtained by solving an energy-balance equation and the conductivity coefficients are then evaluated by carrying out the appropriate averages. The method has been extended to include magnetic fields[21] and applied with apparent success to ionic crystals such as the silver halides.[22]

Since the electron densities in our experiments are orders of magnitude too low for electron-electron inter-

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[22] D. Matz and F. Garcia-Moliner, J. Phys. Chem. Solids **26**, 551 (1965).

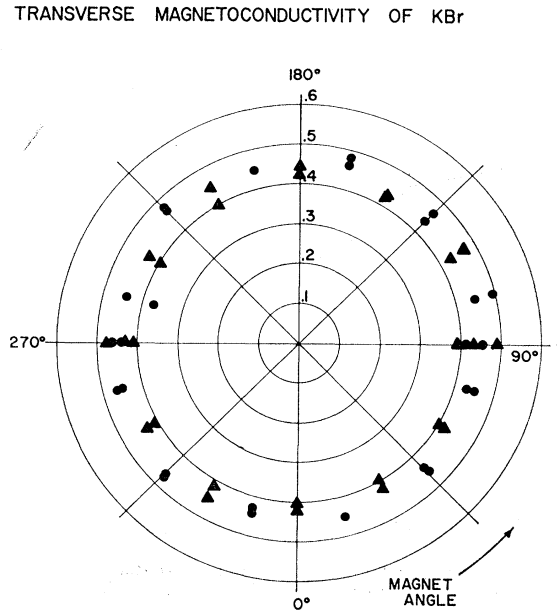

FIG. 7. The effect of a transverse magnetic field as a function of angle. The plot shows the ratio of the currents $i(H)/i(0)$ versus the magnet angle θ for a crystal of KBr at 4.2°K. A magnetic field of 8 kOe is rotated in the x-y or (001) plane of the crystal. An electric field of 112 V/cm was applied in the z direction. The solid dots and triangles represent different runs on the same crystal.

action to be important, the validity of the assumption of a Maxwellian distribution in Shockley's theory is clearly questionable. We, nevertheless, proceed to compare our data with the Shockley model for the following reasons: (1) the more sophisticated theories involve an explicit calculation of the distribution function, a difficult task for mixed scattering mechanisms and high magnetic as well as electric fields, (2) under some conditions, for example, neutral and acoustic scattering and not-too-high field, the transport coefficients are not very sensitive to the exact shape of the distribution function,[20] and (3) we are interested in understanding a variety of fairly involved phenomena and in approximate values for the various important parameters.

Because the details of the Shockley calculation are available in the literature,[20–22] we will only outline the important steps. The shift in mean velocity of the electrons due to electric field is taken into account by a parameter $\gamma=T_e/T_L$, where T_L is the lattice temperature and T_e a "temperature" which characterizes the electron distribution. The parameter γ is obtained by equating the average rate at which electrons gain energy from the electric field $q\mathbf{v}\cdot\mathbf{E}=(q/n)\sigma E^2$ to the rate at which they lose energy to the lattice because of collisions. We are mainly interested in data taken at 4.2°K and low or intermediate electric fields where nonlinearities in current-versus-E begin to set in. For this reason, and for the sake of simplicity, we assume that energy transfer to the lattice occurs mainly through acoustic phonons. The rate of energy loss is therefore given by[21]

$$\left(\frac{d\epsilon}{dt}\right)_{ac}=\frac{32}{3\pi}\frac{qs^2}{\mu_0^{ac}}\gamma^{1/2}(1-\gamma), \tag{4}$$

where $s=3.85\times10^5$ cm/sec, the sound velocity in KBr.[23] An adjustable parameter, the low-field acoustic mobility, enters into Eq (4) and is given by

$$\mu_0^{ac}=\frac{q}{m}\langle\tau_{ac}\rangle=\frac{q}{m}\frac{4}{3\sqrt{\pi}}\tau_0, \tag{5}$$

where τ_0 is a relaxation-time coefficient. When $\gamma\geq1$ the acoustic scattering time is given by $\tau_{ac}=\tau_0(\gamma x)^{-1/2}$, where $x=\epsilon/kT_e$. The computations were carried out using this simple form consistent with equipartition of energy. When this restriction is lifted it can be shown that the experimental results are fitted by somewhat larger values of the parameter μ_0^{ac}.[22]

The conductivity coefficient σ, which enters the energy-balance equation and determines the energy gain from the electric field, is computed from Eq. (3) with a relaxation time involving a combination of mechanisms and a distribution characterized by T_e instead of T_L. The energy-balance equation is then similar to Eqs. (1) and (2) of Ref. 22. Although a combination of acoustic, neutral, dipole, and impurity mechanisms were investigated,[24] the combination of just two scattering probabilities, acoustic and neutral, was most satisfactory. The relaxation time for neutral scattering is given by $\tau=h\tau_0$, where the parameter h can be expressed in terms of a mobility parameter

$$\mu^n=\mu_0^n=\frac{3\sqrt{\pi}}{4}\mu_0^{ac}h.$$

It is this neutral scattering, possibly due to F centers, which mainly determines the observed low-field Hall mobility (approximately 10^4 cm²/V sec) at 4.2°K.[13]

Numerical computations were made for mixed scattering mechanisms with the aid of a high-speed digital computer. For given values of the parameters μ^n, μ_0^{ac}, and field H, the integral involved in the conductivity coefficient was evaluated for some value of γ. This value of γ was then used to check if the energy-balance equation was satisfied; if not, a new value of γ was chosen and the process repeated. Finally, after obtaining γ, the current was evaluated from Eq. (2) with an appropriate value of σ (again an integral similar to Eq. 2 of Ref. 22 was involved). Thus the three solid curves and values of γ shown in Fig. 5 were obtained. These three curves correspond to $H=0$, 18, and 48.7 kOe, respectively, and the very reasonable values of $\mu_0^{ac}=40\,000$ cm²/V sec and $\mu_0^n=15\,200$ cm²/V sec. The fit to the data is quite good for low electric fields, but for high electric fields the observed current in the

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[24] M. Mikkor, thesis, University of Illinois, 1966 (unpublished).

presence of magnetic field $i(H)$ increases and approaches the value of the current without magnetic field $i(0)$ faster than predicted by theory. This is especially noticeable for the case of a magnetic field as high as 48.7 kOe. Notice that the effect of H, besides reducing the conductivity, is to shift the transition from Ohmic to hot-electron behavior to higher electric fields. This is primarily because a transverse magnetic field tends to reduce the rate at which electrons gain energy from the magnetic field.

The isotropic magnetoconductive data of Fig. 7 strongly supports the assumption of standard band shape, at least in the vicinity of $k=0$. It may be that away from $k=0$ nonparabolicity arises due to polaron effects. The energy spectrum of the polaron has been calculated by Whitfield and Puff[25] and more recently by Larsen.[26] The polaron effective mass does tend to increase with k especially as energies comparable to $\hbar\omega_l$ above the bottom of the band are approached. This may have the effect of increasing the acoustic interaction at high fields which would tend to keep the carriers cooler than expected.[27] Thus, even though electrons are strongly coupled to the longitudinal optical mode, LO phonon emission may be relatively unimportant at low or intermediate fields. It is perhaps surprising that the theoretical curves fit as well as they do up to 1000 V/cm and higher (refer to Fig. 5); yet it is conceivable that a smooth transition occurs with increasing field to a region dominated by optical phonon processes.

The present theory indicates that rather high electron temperatures should prevail at fields as high as 1000 V/cm. It is almost certainly true that carriers do not reach these effective temperatures due to polaron effects mentioned above and possibly due to optical phonon emission by electrons in the tail of the distribution. An expression for energy loss through covalent optical modes has been given by Conwell[28] and discussed by Matz and Garcia-Moliner.[21] More recently, energy loss due to polar optical modes has been treated,[29] but it is not clear that these theoretical results can be applied as they stand to the alkali halides where the electron-lattice interaction is so strong. A more general polaron theory has recently been given[30] but has not yet been applied to the case of magnetic fields and mixed scattering.

In spite of the several difficulties which we have emphasized, the energy-balance method appears to be capable of explaining a variety of phenomena at intermediate fields with very reasonable but order-of-magnitude values for μ_0^{ac} and μ^n. These same values, 40 000 and 15 200 cm^2/V sec, respectively, fit the cyclotron-resonance line shape discussed in the following paper.[1] We thus believe that the elementary theory is a reasonable first step toward understanding the general behavior at low or intermediate fields. Certainly a more general theoretical approach capable of giving the actual distribution function, as well as further experiments at high field, are indicated.

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