

state. Thus, this observable overrides the usual energy limitations, and its study may be extended to cover energies from a few eV to tens of MeV.

*Work performed under the auspices of the U.S. Atomic Energy Commission.

†On sabbatical leave to the American University of Beirut, Beirut, Lebanon.

¹M. T. Robinson and O. S. Oen, *Appl. Phys. Letters* **2**, 30 (1960); *Phys. Rev.* **132**, 2385 (1963).

²A. L. Southern, W. R. Willis, and M. T. Robinson, *J. Appl. Phys.* **34**, 153 (1963); L. Lehmann and G. Lieb-fried, *J. Appl. Phys.* **34**, 2821 (1963); G. R. Piercy, M. McCargo, F. Brown, and J. A. Davies, *Can. J.*

Phys. **42**, 1116 (1964); J. A. Davies, G. C. Ball, F. Brown, and B. Domeij, *Can. J. Phys.* **42**, 1979 (1964); O. Almen and G. Bruce, *Nucl. Instr. Methods* **11**, 279 (1961).

³W. Brandt, J. M. Khan, D. L. Potter, R. D. Worley, and H. P. Smith, Jr., *Phys. Rev. Letters* **14**, 42 (1965).

⁴A. F. Tulinov, V. S. Kulikauskas, and M. M. Malov, *Phys. Letters* **18**, 304 (1965).

⁵E. Uggerhoj, *Phys. Letters* **22**, 382 (1966).

⁶J. M. Khan, D. L. Potter, and R. D. Worley, *Phys. Rev.* **148**, 413 (1966).

⁷The hydrogen 1216-Å and the helium 584-Å lines are the strongest of the emission lines of these elements.

⁸R. S. Nelson and M. W. Thompson, *Phil. Mag.* **8**, 1677 (1963).

CYCLOTRON RESONANCE OF THE POLARON IN KCl, KBr, KI, RbCl, AgCl, AgBr, AND TlCl*

J. W. Hodby,[†] J. A. Borders,[‡] and F. C. Brown
Department of Physics, University of Illinois, Urbana, Illinois

and

S. Foner

National Magnet Laboratory,[§] Massachusetts Institute of Technology, Cambridge, Massachusetts

(Received 1 September 1967)

In this Letter we report observations of cyclotron resonance and approximate values for polaron masses in seven different ionic crystals. For the first time, the size of the polaron effect can be estimated in a variety of materials over a wide range of coupling constants. The measurements were carried out at 2-mm wavelengths and magnetic fields up to 140 kOe using an electron-heating technique previously applied to KBr at longer wavelengths.¹ The results can be used to compute band masses (effective masses in the absence of lattice polarization) for the purpose of comparison with recent band calculations.²⁻⁴

Transient currents about 2 μ sec in duration were excited in the insulating crystals studied by repeated flashes of a small xenon flash lamp. The alkali halides were either x rayed or additively colored with *F* centers, and electrons were excited by *K*- and *L*-band illumination. In the case of the silver and thallium halides, the carriers were produced by band-to-band excitation. Identical positive and negative voltage pulses were repetitively applied to the specimen, placed between blocking electrodes immersed in liquid helium. The small transient photocurrents were then amplified by a low-

noise wide-band preamplifier⁵ and pulse-shaping main amplifier. The repeated pulses were averaged in a two-channel boxcar integrating circuit⁶ in such a way that the photoresponse could be continuously recorded as a function of various parameters, for example, as a function of transverse or longitudinal magnetic fields. Polarization effects were minimized by the symmetrical conditions of drift during positive and negative voltage pulses and by the use of a depolarizing light during the dark intervals between pulses. The rms noise of the amplifier was equivalent to a single input pulse of about 450 electrons; however, repeated pulses of only five electrons each could be detected by pulse averaging. Further apparatus details will be given elsewhere.⁷

In order to observe cyclotron resonance of the photocarriers, 140-GHz microwave power from a COE-20C Carcinotron was switched on during alternate pulses of the flash lamp. The output of the boxcar integrating circuit was arranged to be proportional to the difference between alternate photopulses. When using this differential mode, microwave-induced changes in the photoresponse as small as 0.1% could be observed. The crystal was located

in a longitudinal magnetic field as in Ref. 1, except that a water-cooled Bitter-type solenoid was employed instead of a superconducting solenoid. The Carcinotron was located about 10 ft away from the magnet in order to avoid fringing field effects. Transmission through cylindrical TE_{01} mode waveguide minimized losses. Rapid switching of the microwaves was achieved either by the use of a ferrite modulator or by frequency modulation of the Carcinotron over the passband of the rectangular-to-cylindrical waveguide mode converter. The high differential sensitivity of the detection apparatus permitted the use of lower microwave field strengths than in Ref. 1, consequently, large untuned samples were employed rather than resonant dielectric posts.¹

Samples of the various alkali halides were selected for large $\omega\tau$ by independent transport experiments, either by observing a low-temperature Hall mobility in excess of 10^4 cm²/V sec, or by observing a critical field (onset of hot-electron behavior) in the vicinity of 100 V/cm.^{1,8} For KCl, KBr, KI, and RbCl as well as NaI and CsBr, the transverse magnetconductivity was measured by maintaining the magnitude of the field constant, but allowing its direction to rotate through 360° in a plane orthogonal to the applied electric field. This was done at various electric field strengths above and below the critical field, and no variations

greater than 0.5% were observed which were consistent with cubic symmetry. These results are in agreement with the assumption of spherically symmetric conduction bands centered at $k=0$ as well as with isotropic relaxation times. Similar results were found for electrons in the silver halides by Tippins.⁹

Using the differential technique described above, definite evidence for cyclotron resonance was obtained for the materials listed in Table I. A single resonance was found in each case except for TlCl, where both a low- and a very high-field resonance was observed. Figure 1 shows the resonance at $H=51$ kOe for electrons excited from F centers in KBr. It should be pointed out that the sign and magnitude of the differential resonance signal shown in Fig. 1 is associated with a small decrease (0.5%) in the direct current mobility as electron energy is increased by the application of microwave power. The differential signal can also be of opposite sign under conditions where the electron mobility tends to increase with electron energy. Such effects were sometimes observed at low electric fields and were associated with superlinearity in current versus voltage curves taken under the conditions of the experiment. This effect could arise from a transverse component of magnetic field due to misalignment (by 1° or 2°) between applied electric and magnetic fields. Transport effects in the alkali

Table I. Observed (polaron) mass m_p , band mass m , and coupling constant α for the materials shown. The high (ϵ_∞) and low (ϵ_s) frequency dielectric constants and longitudinal optical frequencies $1/\lambda$ are also given. The last two columns contain the mass m^* from band theory and reduced masses $\mu = \epsilon_\infty^2 G/13.6$ from exciton optical data.

	m_p/m_e	m/m_e	α	ϵ_∞	ϵ_s^a	$1/\lambda_l^a$ (cm ⁻¹)	m^*/m_e	μ/m_e
KCl	1.25 ± 0.12	0.496	3.97	2.13	4.49	213	0.55^b	0.13^d 0.67^e
KBr	0.93 ± 0.05	0.428	3.52	2.42	4.52	168		
KI	0.67 ± 0.05	0.398	2.60	2.76	4.66	143	0.49^c	0.22^f
RbCl	1.38 ± 0.10	0.515	4.14	2.22	4.58	180		
AgCl	0.51 ± 0.04	0.348	2.00	4.04	9.50	197		
AgBr	0.33 ± 0.03	0.241	1.69	4.62	10.6	140		
TlCl	0.53 ± 0.05	0.320	2.55	5.10	37.6	158		
	2.72 ± 0.10	0.936	4.35					

^aR. P. Lowndes, Phys. Letters 21, 26 (1966).

^bRef. 3.

^cRef. 2.

^dFor $h\nu_2=8.4$ [T. Tomiki, J. Phys. Soc. Japan 21, 409 (1966); see also G. Baldini and B. Bosacchi, to be published], $E_0=8.5$ eV [J. Phillips, Phys. Rev. 136, A1705 (1964)], and the use of ϵ_∞ .

^eFor $h\nu_2=8.4$ [T. Tomiki, J. Phys. Soc. Japan 21, 409 (1966); see also G. Baldini and B. Bosacchi, to be published], $E_0=8.9$ eV [G. Huggett and K. Teegarden, Phys. Rev. 141, 797 (1966)], and the use of ϵ_∞ .

^fJ. Ramamurti and K. Teegarden, Phys. Rev. 145, 698 (1966).

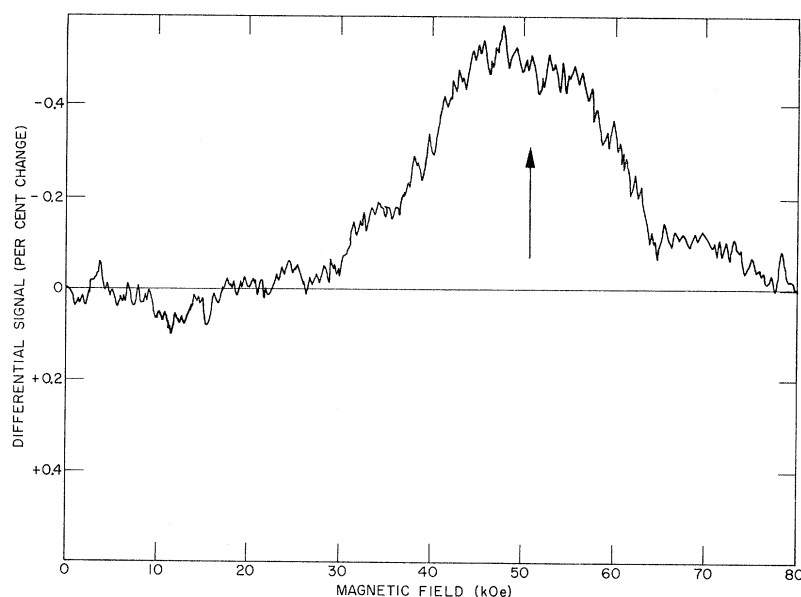


FIG. 1. Cyclotron resonance signal at 140 GHz for photoelectrons in an additively colored crystal of KBr at 4.2°K. The vertical scale is inverted and approximately gives the percent change in photocurrent produced by the microwaves. This is a single scan. Repeated scans show that the maximum effect occurs in the vicinity of 51 kOe corresponding to a polaron mass $m_p = eH/\omega c = 0.93m_e$.

halides were also observed which could be associated with spin resonance of F centers. These will be reported elsewhere.

The 51-kOe peak shown in Fig. 1 corresponds to a cyclotron-resonance effective mass of 0.93 ± 0.05 free-electron masses. Presumably this measured mass (spherical energy surfaces) is close to that of the polaron since the longitudinal optical frequency in KBr (5100 GHz)¹⁰ is much higher than the cyclotron frequency at resonance (140 GHz). Effects such as discussed by Johnson and Larsen¹¹ are not expected to be very important in the present experiments. We therefore use the symbol m_p for both observed and polaron mass since they are probably very nearly the same number.

The various observed polaron masses, m_p , are given in the first column of Table I, whereas the second column contains values of band mass, m , computed by means of an interpolation formula due to Langreth.¹² This formula, which closely approximates the Feynman polaron mass, is

$$m_p = m \left(\frac{1 - 0.0008\alpha^2}{1 - \frac{1}{6}\alpha + 0.00034\alpha^2} \right), \quad (1)$$

where the coupling constant $\alpha = (e^2/\hbar)(m/2\hbar\omega_l)^{1/2} \times (\epsilon_s - \epsilon_\infty)/\epsilon_s\epsilon_\infty$, ϵ_s is the static dielectric constant, ϵ_∞ the optical dielectric constant, and

ω_l the longitudinal optical frequency. It is interesting to compare these band masses m with the predictions of band theory as shown for two cases^{2,3} in the next to the last column of Table I. Reasonable agreement is found.

The exciton lines of the alkali halides appear to fit approximately a Wannier model and series of the form $h\nu_n = E_0 - G/n^2$, where E_0 is the interband transition energy and G is the exciton binding energy.¹³ Dielectric-constant data and the exciton optical values for G can therefore be used to deduce a reduced exciton mass $\mu = \epsilon^2 G / 13.6$, which should be close to the effective mass of the electron for the narrow valence bands of the alkali halides. Because of the local nature of the $n = 1$ exciton state, it seems most appropriate to determine G from E_0 and $h\nu_2$ or higher terms in the series when these energies are known. A favorable case is that of KI, where $G \approx 0.4$ eV.¹⁴ Now in determining μ from the above formula, there is a difficulty as to the proper choice of dielectric constant. The use of the optical dielectric constant for KI, $\epsilon_\infty = 2.76$, gives $\mu/m_e = 0.22$. This value of μ is considerably less than the band mass shown in the second column of Table I. It thus appears that an effective dielectric constant larger than ϵ_∞ must be used for these fairly large orbit exciton states. A value of

$\epsilon = 3.7$, intermediate between ϵ_∞ and ϵ_s , gives a reduced mass μ close to the electron band mass m . When the static constant $\epsilon_s = 4.66$ is employed, a reduced mass $\mu/m_e = 0.64$ is obtained which is close to the polaron mass for KI given in the first column of Table I. Such a result is consistent and would be expected for excitons of sufficiently large radius as discussed by Haken.¹⁵ The question is whether or not bound polaron states with the binding characterized by an effective dielectric constant as large as ϵ_s are produced during the absorption process itself.¹⁸

Recently, transitions to the $n=2$ exciton states have been identified in the reflection spectra of KCl and other alkali halides.¹⁷ On the other hand, values of the band gap energy E_0 are not yet known with precision. Table I lists the reduced masses in KCl using ϵ_∞ and two choices for E_0 .^{18,19} The first choice, corresponding to a smaller binding energy, yields a value of μ much less than the value of m given in the second column. Again, this would indicate that one should employ an effective dielectric constant somewhat larger than ϵ_∞ as in the case of KI.

Cyclotron resonances were also found for band-to-band excitation in AgBr, AgCl, and TlCl. In the first two cases these are almost certainly due to electrons in agreement with a Hall effect of negative sign in the silver halides. The resonance for AgBr overlaps, but is a little higher than, that previously reported.²⁰ Plasma resonance effects, which are certainly absent here, may have contributed to the earlier result at higher temperatures and high light levels. The smaller of the two masses for TlCl is in close agreement with recent cyclotron resonance absorption measurements at 6 mm.²¹ Both electrons and holes have been found to be mobile in TlCl.²²

It is a pleasure to acknowledge helpful discussions with G. F. Dresselhaus, M. E. Dresselhaus, B. Lax, and D. M. Larsen. The authors also appreciate the help of L. G. Rubin, B. Thaxter, and R. Brandt. Funds from an Eastman Kodak grant at the University of Illinois are gratefully acknowledged.

*Work supported in part by the U. S. Army Research Office (Durham) under Contract No. ARO(D)-217.

†Permanent address: Clarendon Laboratory, Parks Road, Oxford, England. Part of this work was performed while the author was a Guest Scientist at National Magnet Laboratory.

‡In partial fulfillment of the Ph. D. requirements at the University of Illinois. Part of this work was performed while the author was a Guest Scientist at the National Magnet Laboratory.

§Supported by the U. S. Air Force Office of Scientific Research.

¹M. Mikkor, K. Kanazawa, and F. C. Brown, *Phys. Rev. Letters* **15**, 489 (1965); also, *Phys. Rev.* (to be published).

²Y. Onodera, M. Okayaki, and T. Inui, *J. Phys. Soc. Japan* **21**, 816 (1966).

³P. De Cicco, *Phys. Rev.* **153**, 931 (1967).

⁴S. Oyama and T. Miyakawa, *J. Phys. Soc. Japan* **21**, 868 (1966).

⁵Type TC-130, Tennelec Instrument Company, Oak Ridge, Tennessee.

⁶R. J. Blume, *Rev. Sci. Instr.* **32**, 1016 (1961).

⁷J. Borders and J. Hodby, to be published.

⁸F. Nakazawa and H. Kanzaki, *J. Phys. Soc. Japan* **22**, 844 (1967).

⁹H. H. Tippins and F. C. Brown, *Phys. Rev.* **129**, 2554 (1963).

¹⁰R. P. Lowndes, *Phys. Letters* **21**, 26 (1966).

¹¹E. J. Johnson and D. M. Larsen, *Phys. Rev. Letters* **16**, 655 (1966).

¹²D. C. Langreth, *Phys. Rev.* **159**, 717 (1967).

¹³F. Fischer, *Z. Physik* **160**, 194 (1960).

¹⁴J. Ramamurti and K. Teegarden, *Phys. Rev.* **145**, 698 (1966).

¹⁵H. Haken, *Z. Physik* **155**, 223 (1959).

¹⁶R. S. Knox and K. J. Teegarden, in *The Physics of Color Centers*, edited by W. B. Fowler (Academic Press, Inc., New York, to be published).

¹⁷T. Tomiki, *J. Phys. Soc. Japan* **21**, 409 (1966); see also, G. Baldini and B. Bosacchi, to be published.

¹⁸J. C. Phillips, *Phys. Rev.* **136**, A1705 (1964). Recent two quantum experiments also seem to favor the smaller band gaps and binding energies. C. D. Fröhlich and B. Stagginnus, *Phys. Rev. Letters* **19**, 496 (1967).

¹⁹G. Huggett and K. Teegarden, *Phys. Rev.* **141**, 797 (1966); see also G. Baldini and K. Teegarden, *J. Phys. Chem. Solids* **27**, 943 (1966).

²⁰G. Ascarelli and F. C. Brown, *Phys. Rev. Letters* **9**, 209 (1962).

²¹T. Masumi and H. Tamura, private communication.

²²T. Kawai, K. Kobayashi, and H. Fujita, *J. Phys. Soc. Japan* **21**, 453 (1966).