(1971).

[3] R. Zallen, M. L. Slade, and A. T. Ward, Phys. Rev. B 3, 4257 (1971).

[4] G. Fischer and J. L. Brebner, J. Phys. Chem. Solids 23, 1363 (1962).

[5] P. C. Leung, G. Andermann, W. G. Spitzer, and C. A. Mead, J. Phys. Chem. Solids 27, 849 (1966).

[6] F. Jellinek and H. Hahn, Z. Naturforsch. 16b, 713 (1961).

[7] R. W. G. Wyckoff, *Crystal Structures* (Interscience, New York, 1965), 2nd ed., Vol. 1, p. 145.

[8] D. F. Hornig, J. Chem. Phys. 16, 1063 (1948); E. B. Wilson, Jr., J. C. Decius, and P. C. Cross, *Molecular Vibrations* (McGraw-Hill, New York, 1955), Appendix X, pp. 312–340.

[9] In previous papers we have used the term "quasi-acoustical" instead of "rigid-layer." However, the latter term is perhaps clearer and more descriptive, since in the long-wavelength limit there is no relative displacement of the atoms within the layers.

[10] The GaSe crystal was grown by J. P. Voitchovsky of the École Polytechnique Fédéral de Lausanne, Switzerland.

[11] H. O. McMahon, J. Opt. Soc. Am. 40, 376 (1950).

[12] M. Born and K. Huang, *Dynamical Theory of Crystal Lattices* (Oxford U. P., Oxford, England, 1968), Sec. 9.

[13] G. B. Wright and A. Mooradian [Bull. Am. Phys. Soc. 11, 812 (1966)] have observed Raman lines in GaSe at 59.6, 133.8, 209.5, 253.8, and 308.6 cm^{-1}. However, no polarization properties were given, and the low-frequency mode at 19.1 cm^{-1} was not reported.

[14] K. R. Symon, *Mechanics* (Addison-Wesley, Reading, Mass., 1957), p. 165.

[15] L. Pauling, *The Nature of the Chemical Bond* (Cornell U.P., Ithaca, New York, 1960), p. 257.

[16] R. A. Bromley, Phil. Mag. 23, 1417 (1971).

PHYSICAL REVIEW B VOLUME 5, NUMBER 4 15 FEBRUARY 1972

Experimental Observation of Wannier Levels in Semi-Insulating Gallium Arsenide[†]

Robert W. Koss
University of Vermont, Burlington, Vermont

and

L. M. Lambert*
Norwegian Institute of Technology, Trondheim, Norway

(Received 3 August 1971)

Optical absorption in an electric field has been of increased interest in recent years since Callaway predicted that the Wannier levels may be observable in direct-transition semiconductors such as GaAs. In this work, such levels have been observed for the first time and found to be in substantial agreement with the Callaway theory.

INTRODUCTION

This paper reports on an experimental investigation of the effect of a uniform external electric field on optical absorption in semi-insulating GaAs. Experimental data were obtained from room temperature to 24 °K for electric fields up to 1.6×10^5 V/cm. The existence of Wannier levels was clearly evident at 24 °K and agreed well with the Callaway theory.

The direct-transition absorption coefficient predicted by Callaway[1] using Kane functions was given as

$$\alpha = \frac{4\pi^2 \kappa \mu \beta^{2/3}}{\omega K} \sum_{j=j_0}^{\infty} A_i^2 \left(\frac{2\pi j \beta^{1/3}}{K} - \chi \right) , \qquad (1)$$

where

$$\kappa = \frac{2e^2 \,|\, \vec{\xi} \cdot \vec{p}_{nn'} |^2}{\pi \hbar m^2 n \epsilon_0 c} , \quad \chi = \tfrac{1}{12} K^2 / \beta^{2/3} , \quad \beta = 2\mu F / \hbar^2 ,$$

and where $A_i(x)$ is the usual Airy function, ω is the photon frequency, K is the width of the Brillouin zone along a principal lattice direction of the applied field, F is the electric field force, μ is the reduced effective mass, $\vec{p}_{nn'}$ is the interband-momentum matrix element, $\vec{\xi}$ is the photon polarization vector, and n is the index of refraction.

The lower limit of the summation is given by j_0, where j_0 is dependent upon the photon energy $\hbar\omega$, and is the next integer greater than q_0, where q_0 is given by

$$q_0 = \frac{K}{2\pi F} \left(\frac{\hbar^2 K^2}{24\mu} + E_g - \hbar\omega \right). \qquad (2)$$

To increase the value of q_0 by 1, it is necessary to decrease the photon energy by

$$\Delta\hbar\omega = 2\pi F / K . \qquad (3)$$

Thus, when α is plotted as a function of increasing photon energy for a given electric field, the result is a monotonically increasing staircase. The width of each step is proportional to the electric field as given by Eq. (3).

For small, uniform electric fields, theory predicts an average change $\Delta\alpha$ as

$$\Delta\alpha = \frac{2\pi K \mu \beta^{1/3}}{\omega} \left[\left(\frac{dA_i(y)}{dy} \right)^2_{y=y_0} - y_0 A_i^2(y_0) \right] , \tag{4}$$

where y is the argument of the Airy function in Eq. (1) and y_0 is the value of this argument for $j = q_0$.

Equations (1) and (4) are directly applicable to a two-band direct transition (i.e., single valence and conduction bands). In applying these results to real semiconductors with degenerate valence bands of different effective masses, a weighted average of both bands proportional to their density of states must be used as

$$\alpha = \frac{\alpha_1 \mu_1 + \alpha_2 \mu_2}{\mu_1 + \mu_2} , \tag{5}$$

where α_1 and α_2 are the two-band transitions between each valence band and the single conduction band with the corresponding reduced masses μ_1 and μ_2. Equation (5) is based essentially on two assumptions, both of which are valid for GaAs in this experiment. First, we have assumed that the independent-particle approximation holds; i.e., the energy of the hole-electron pair in the electric field is much larger than the maximum exciton binding energy. For the exciton levels in GaAs and for fields of 10^5 V/cm or greater, this condition is well satisfied.[2] Second, we assumed that the bands are independent; i.e., one band is not filled or depleted by transport processes in the high electric field during optical absorption. Again for low photon fluxes this condition is satisfied in semi-insulating GaAs.[3]

It should be noted in Eq. (1) that both the argument and phase shift of the Airy function are functions of reduced mass. Thus one finds that the "staircase" function for the light-hole–conduction-band transition is shifted in energy and increased in amplitude with respect to the heavy-hole–conduction-band transition. The result is a rather complex staircase as shown in Fig. 1.

The effect of the averaging process of Fig. 1 is significant, inasmuch as the resulting Wannier

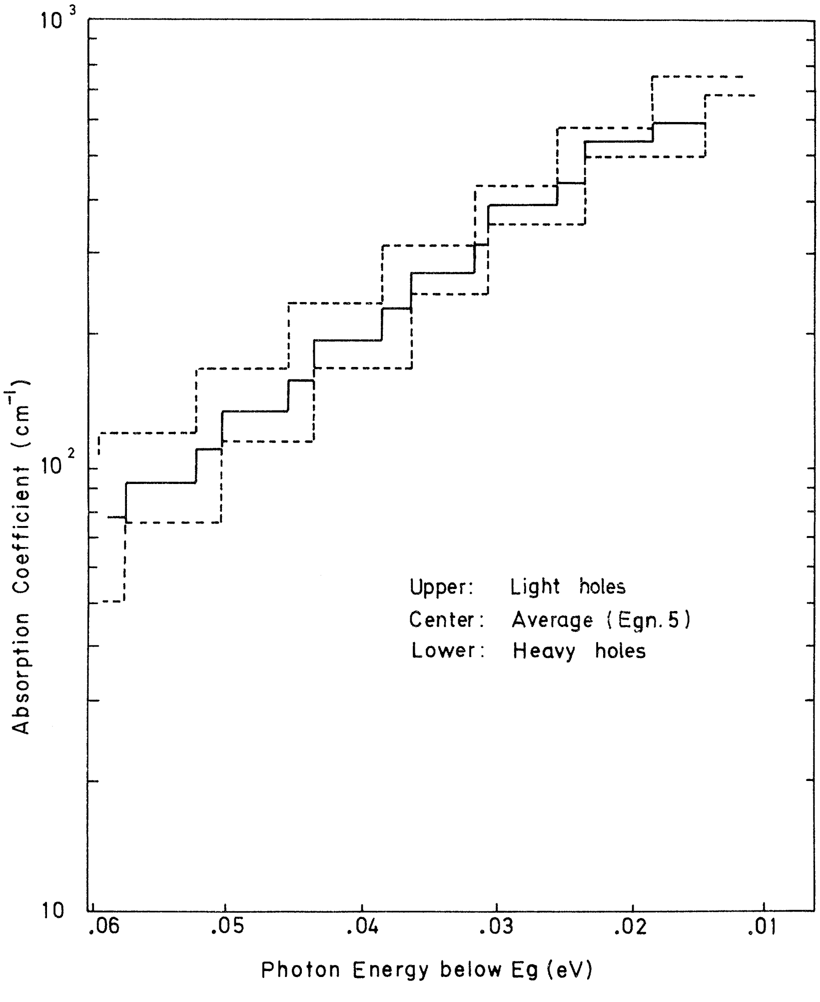

FIG. 1. Calculated Wannier levels for [111] GaAs $E = 1.2 \times 10^5$ V/cm.

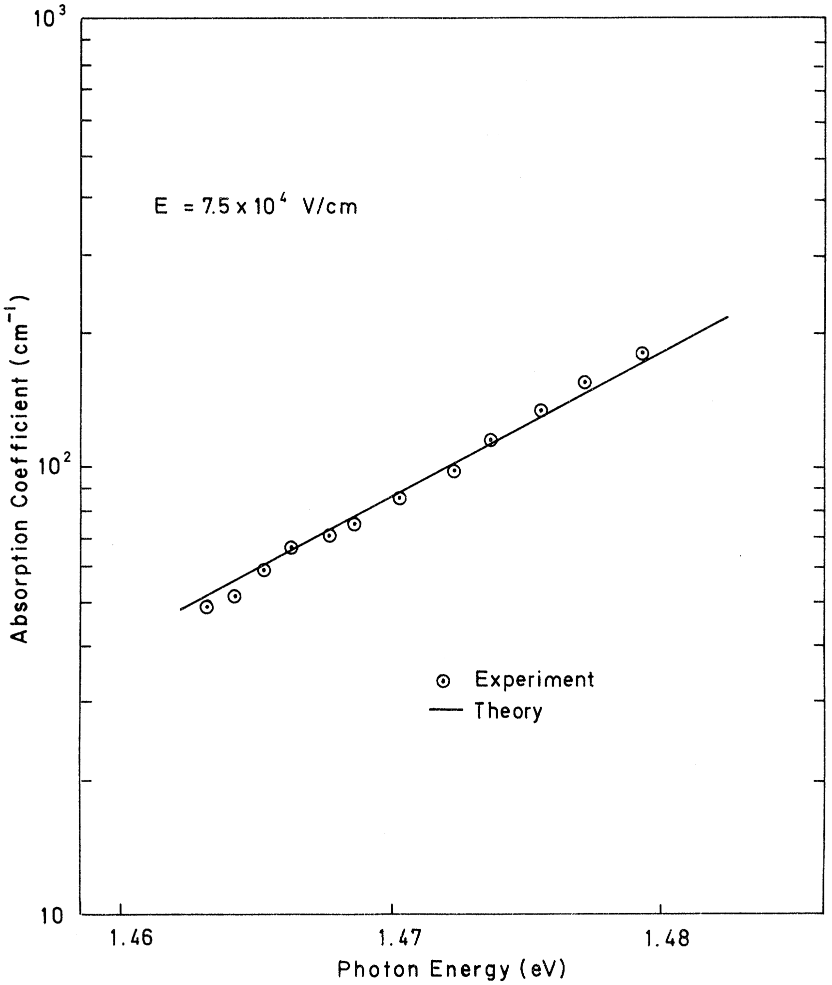

FIG. 2. Electric field absorption coefficient at 24 °K (Franz-Keldysh effect).

staircase has a sequence of intermediate steps. Further, the widths of the resultant steps are not linear in electric field as given in Eq. (3).

EXPERIMENTAL PROCEDURES

The experimental procedures used were essentially the same as reported previously.[4] The electric field effect for optical absorption was measured at selected photon energies less than the gap energy with an over-all energy resolution equal or greater than 10^{-3} eV for all data points reported here.

The samples were semi-insulating GaAs having a resistivity in excess of 10^8 Ω cm at room temperature and oriented with the [111] direction normal to the surface. Silicon monoxide was used as the insulator for blocking contacts.

A simple capacitance measurement at 150 kHz was used to investigate the change in the sample package with temperature and thus give the relationship between applied voltage and the electric field in the GaAs. The result was confirmed by comparison with known low-temperature Franz-Keldysh effect data.[4–6] However, because of the complexity of the sample package, we estimate that an error of no more than ± 5% can exist in determining the actual electric field in the GaAs. This error, though small, is significant; therefore, we used the electric field as obtained from the experimental Franz-Keldysh effect as the correct value for calculation of the Wannier staircase.

EXPERIMENTAL RESULTS

The existence of Wannier levels between 77 and 300 °K was sought and found to be absent as they should be because of thermal broadening of the levels. Certain samples, however, exhibited oscillations in optical absorption throughout this whole temperature range. Such oscillations appeared to be temperature independent, but were sample dependent (i.e., larger in some samples than others). Similar oscillations appear to have been observed by French at 90 °K in GaAs,[5] and by Vavilov *et al.* at 77 °K in CdS.[7] Samples exhibiting this type of oscillation strongly were not used in this investigation.

Data were taken between liquid-helium and liquid-nitrogen temperatures for fields up to 1.6×10^5 V/cm.

The usual procedure (4) of making a relative measurement of absorption coefficient on an expanded scale in order to achieve maximum accuracy and then calculating the absolute absorption for purposes of plotting these figures was used.

In Fig. 2, the field is sufficiently small so that only the Franz-Keldysh effect is evident. The linear fit to the data points determined the relationship between applied voltage and electric held in the GaAs. The slight oscillations observed are real and are typical of the sample-dependent type cited earlier.

The onset of the Wannier levels becomes quite evident at fields in excess of 10^5 V/cm as seen in Fig. 3. The effect is small even at these fields but the data were reproducible. The maximum estimated error for each data point is as shown. The instrument dispersion and slit width dictated the energy error, which is admittedly large; however, the measured absorption error is small, since a time average of each point was obtained to reduce the noise. Both the phase and amplitude of theory appear to be well confirmed by the experimental data.[8] Below 1.45 eV in Fig. 3 for the smallest field (curve 1) the long-period sample-dependent oscillation is clearly evident.

These results do not agree with that of French[5] and Vavilov *et al.*[7] for several reasons. First, if Eq. (3) is correct, it is clear that the Wannier levels should not be observable above 40 °K, which these were not. The oscillations observed by French and Vavilov *et al.* were much larger than the calculated Wannier levels and appear to be similar to the sample-dependent oscillations observed by us, which were temperature independent. Secondly, the accuracy of measurement requires that fields in excess of 10^5 V/cm be used to obtain reliable data. For the experimental arrangement used by Vavilov *et al.* for CdS, an even larger peak field would be necessary, since they were measuring

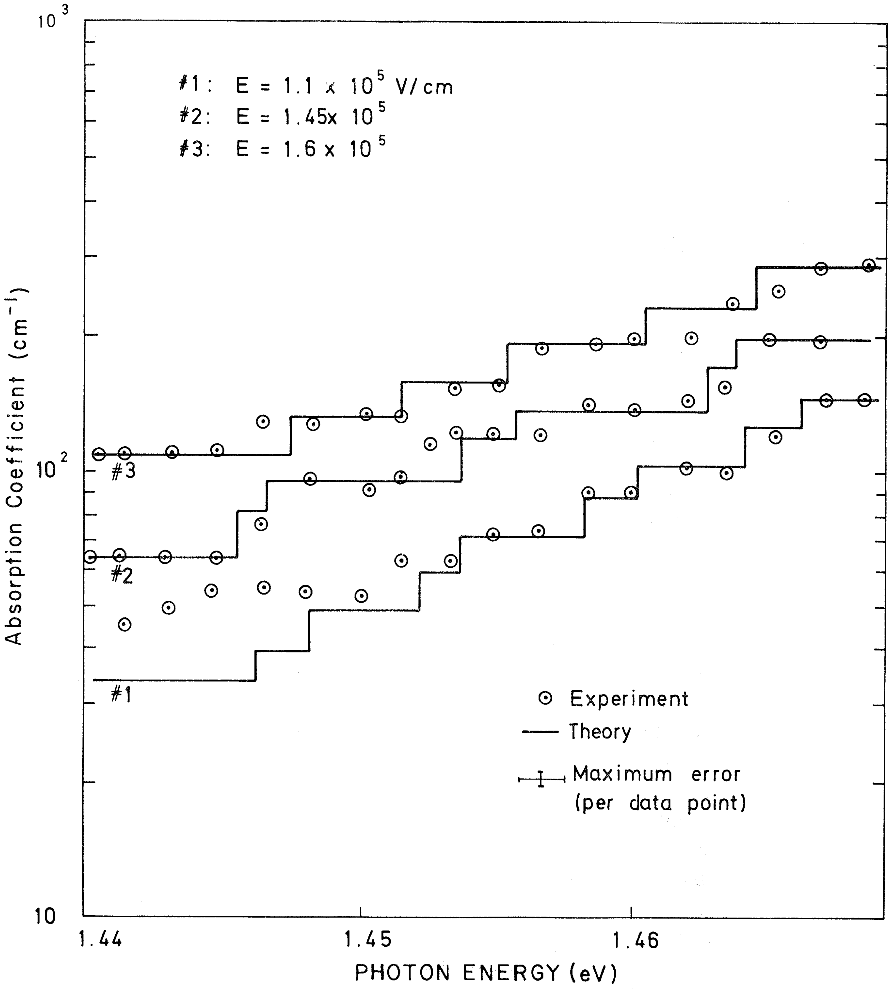

FIG. 3. Electric field absorption coefficient at 24 °K (Wannier levels).

rms absorption. There is also doubt that Wannier levels can be observed in CdS, owing to the large exciton binding energies.[2] Finally, since a time average of data must be taken at these small $\Delta\alpha$ levels in order to eliminate noise, heating of the sample can cause serious error unless the field is turned off except during the actual measurement time.[4] Errors of this latter type increase the vertical error bar dramatically through the temperature dependence of E_g.

CONCLUSIONS

The results of these experiments clearly indicate the existence of very small field- and temperature-dependent oscillations. These oscillations agree well with the Callaway theory for Wannier levels if the light- and heavy-hole transitions are considered to be independent and if the results are averaged proportional to the effective masses (density of states) of each band.

In addition to the small Wannier oscillations, longer-period temperature-independent and sample-dependent (for amplitude) oscillations were also observed. It is believed that these oscillations are those observed by French[5] and Vavilov *et al.*[7]

---

†Work supported in part by the U. S. Air Force, under Grant No. AF-AFOSR-1157-66 and NASA institutional grants; done in partial fulfillment of the requirements for the Ph.D. by R. W. Koss at the University of Vermont.

*Permanent address: University of Vermont, Burlington, Vt.

[1] J. Callaway, Phys. Rev. 130, 549 (1963).

[2] For an analytical treatment see, e.g., C. B. Duke and M. E. Alferieff, Phys. Rev. 145, 583 (1966); H. I. Ralph, J. Phys. C 1, 378 (1968). For the case where excitons cannot be ignored (i.e., in CdS) see L. M. Lambert, Bull. Am. Phys. Soc. 14, 248 (1969).

[3] For a discussion of this specifically for *p*-type Ge see W. E. Pinson and R. Bray, Phys. Rev. 136, A1449 (1964).

[4] L. M. Lambert, J. Phys. Chem. Solids 26, 1409 (1965); Phys. Rev. 138, A1569 (1965).

[5] B. T. French, Phys. Rev. 174, 991 (1968).

[6] E. G. S. Paige and H. D. Rees, Phys. Rev. Letters 16, 444 (1966).

[7] V. S. Vavilov, V. B. Stopachinskii, and V. Sh. Chanbarisov, Fiz. Tverd. Tela 8, 2660 (1966) [Sov. Phys. Solid State 8, 2126 (1967)].

[8] The effective masses for GaAs proposed by R. W. Shaw [Phys. Rev. B 3, 3283 (1971)] were used for these calculations.

PHYSICAL REVIEW B VOLUME 5, NUMBER 4 15 FEBRUARY 1972

Low-Temperature Scattering in InSb Measured by Infrared Faraday Rotation*

T. O. Poehler and C. H. Wang
Applied Physics Laboratory, The Johns Hopkins University, Silver Spring, Maryland 20910

(Received 16 September 1971)

Low-temperature electron-impurity scattering has been measured in *n*-type InSb using both far-infrared Faraday rotation and ellipticity. The experimental results at 5 °K using 337- and 119-μm radiation yielded scattering times of $3-5\times10^{-12}$ sec, which are in good agreement with earlier measurements of scattering using cyclotron resonance linewidths. The results show that dc theory cannot be used to describe this experimental regime, but must be replaced by a more complete frequency-dependent theory of Coulomb interactions.

INTRODUCTION

The low-temperature dc conductivity of compound semiconductors, such as InSb, is governed by phonon scattering down to about 60 °K, while below this temperature ionized impurity scattering dominates. One might expect these mechanisms to operate at infrared wavelengths, since measurements of scattering times in some materials at high microwave frequencies have yielded the same values as deduced from dc measurements.[1,2] However, recent measurements of low-temperature electron-impurity scattering times in compound semiconductors using millimeter and infrared cyclotron resonance linewidths have yielded scattering times greatly in excess of those determined by dc mobility measurements.[3-5] It would appear desirable to make independent determinations of these scattering times which are not necessarily related to a resonant process for comparison with the results of the linewidth measurements. The free-carrier Faraday effect provides an excellent method for measuring low-temperature scattering over a wide range of magnetic fields. The Faraday effect has been widely used to measure the effective mass of free carriers in many compound semiconductors.[6] Use