

Letters to the Editor

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Compacted Scintillators

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AN investigation has been made of the properties of scintillators prepared by high-pressure compaction in an evacuated die using techniques similar to those employed in infrared analysis.¹ A number of representative phosphor systems have been examined to date including NaI(Tl), KBr(Tl), anthracene and ZnS(Ag). In all cases, activated microcrystalline powders were subjected to compaction pressures of the order of 100 000 psi in a small double-piston die from which the air had been withdrawn by a mechanical vacuum pump. Pressures were held for periods from 5 to 15 minutes and then the die opened, and the material removed. Precautions were taken with hygroscopic substances such as NaI(Tl) to avoid water vapor during the entire procedure.

In successful compactations reasonably clear cylindrical masses are obtained with diameters of ~ 1.3 cm and thicknesses of ~ 0.4 cm. Such results have been obtained thus far for both NaI(Tl) and KBr(Tl) although some light scattering within the compacted volume is still evident. It seems likely that the residual scattering will be greatly reduced with better dies operating at higher effective pressures and with more care in the preparation of the microcrystalline powders. Results have not been as promising with anthracene which, presumably as a result of chemical reaction under pressure, compacts to an almost opaque, highly discolored substance. ZnS(Ag) compacts in separated, somewhat translucent layers at the piston surfaces with the majority of the mass remaining powdery. Again more efficient dies operating at higher effective pressures than our present die may produce better results.

For compacted NaI(Tl) and KBr(Tl), the scintillation efficiency was determined by direct pulse-height comparison with single-crystal samples² of similar size using a DuMont K 1186 PM tube and Cs¹³⁷ gamma rays. For KBr(Tl) we consistently obtain ninety percent of the single crystal pulse height while for NaI(Tl) we have obtained values ranging from 35 percent to 85 percent.

The spread for NaI(Tl) is due primarily to the inadequacy of our techniques for handling the microcrystalline powder. No detailed measurements have been made on scintillation decay time, but any difference between the single crystals and the compacted material is certainly slight.

The comparatively high efficiency of properly prepared compacted scintillators indicates that the plastic flow and lattice destruction accompanying the process does not interfere seriously with the gross effectiveness of energy transfer from the host crystal to the activator sites. Furthermore, it seems likely that afterglow phenomena associated with deep traps will be less pronounced with unannealed compacted scintillators than with single crystals. No attempts at annealing or other treatments to increase the scintillation pulse height have been made as yet.

It appears possible that compaction techniques may ultimately provide effective means for producing large sensitive volumes while retaining most of the advantages of present inorganic scintillators. It is also possible that greater control of activator distribution will be obtainable, making practical such highly concentration-sensitive systems as LiI(Tl).³

¹ M. M. Stimson, *J. Chem. Soc.* **74**, 1805 (1952); U. Schiedt and H. Reinwein, *Z. Naturforsch.* **7B**, 270 (1952).

² Scintillation grade material obtained from the Harshaw Chemical Company.

³ S. C. Curran, *Luminescence and the Scintillation Counter* (Academic Press, Inc., New York, 1953), p. 133.

Infrared Absorption of Germanium near the Lattice Edge

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WE have noticed that the absorption spectrum of a number of semiconductors, PbS, PbSe, PbTe, Mg₂Sn, and Ge obeys a law of the form $K \propto (h\nu - E_0)^n$, where n is either 2 or 3, at low values of the absorption constant K near the long wave edge of the main lattice absorption band. It appeared to us that such laws could be understood if the tail of the lattice edge absorption arose from optically "forbidden" transitions and that the energy E_0 would then be closely related to the minimum energy gap.

In a recent letter Hall, Bardeen, and Blatt¹ have calculated the optical absorption by valence electrons for "indirect" and "direct" transitions. For the "indirect" transitions they give a law of the form $K \propto (h\nu - E)^2 \{k_e^2 + \hbar^{-2}(m_c + m_v)(h\nu - E)\}$, which is in agreement with our observations. In view of the qualitative success of the theory it seemed to us important to obtain sufficient data to test it quantitatively, and for this purpose we have chosen to study germanium. As

published data on absorption in germanium² is inadequate for our purpose, we have measured the absorption for a 60 ohm cm polycrystalline specimen using glass prisms to get the highest resolution. The method depends essentially on measuring the radiation intensity with and without the specimen present, deducing the reflection coefficient, R , from the constant value of the transmission coefficient at wavelengths well beyond the absorption edge, and using the measured thickness of the germanium plate to calculate the absorption constant. Due allowance was made for multiple reflections.² R was found to vary slightly with temperature as given in Table I. It should be noted that the value of R at

TABLE I. Dependence of reflection coefficient on temperature.

T°K	291	249	195	90	77	20	4.2
R	0.382	0.379	0.375	0.373	0.370	0.369	0.368

room temperature is higher than the one obtained by Briggs,³ *viz.*, 0.365. Absorption curves were taken at seven temperatures ranging from 4.2°K to 291°K. We have found that K can be well represented at low levels of absorption by a law of the form

$$K = A \left[\frac{1}{1 - e^{-\theta/T}} \left(\frac{h\nu - E_G - k\theta}{h\nu} \right)^2 + \frac{1}{e^{\theta/T} - 1} \left(\frac{h\nu - E_G + k\theta}{h\nu} \right)^2 \right]. \quad (1)$$

Thus when $K^{1/2}$ is plotted against $h\nu$, as in Fig. 1, the points lie close to one straight line for $E_G - k\theta < h\nu < E_G + k\theta$ to a steeper straight line for $h\nu > E_G + k\theta$. This behavior is just what one expects if the absorption in this range is due to indirect transitions in which there is a marked change in momentum between the initial and the final states and in which direct transitions do not play a role. The first term in brackets then refers to

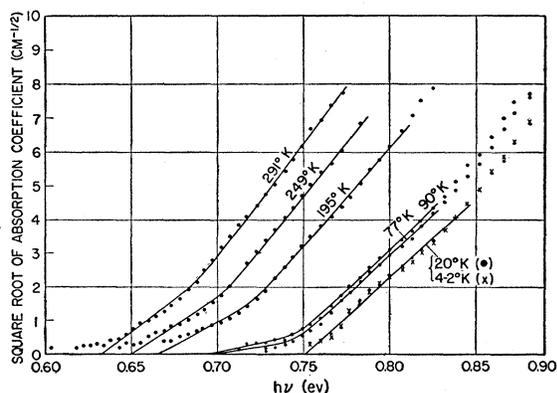


FIG. 1. Dependence of absorption constant on photon energy for 60 ohm-cm germanium.

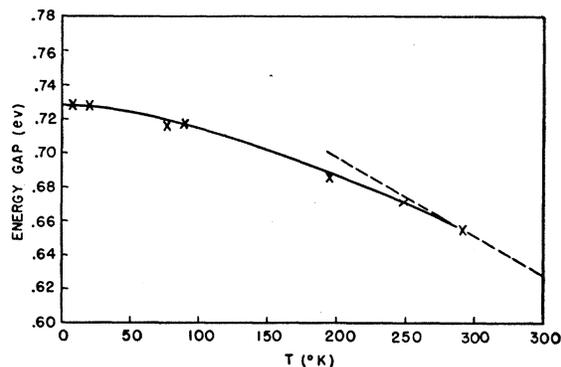


FIG. 2. Dependence of energy gap on temperature for germanium.

photon absorption with emission of a phonon of energy $k\theta$, the second to photon absorption with absorption of a phonon of energy $k\theta$, and E_G is the minimum energy gap. We find that if $\theta = 260^\circ\text{K}$ the dependence of K on $h\nu$ and on T follows closely the prediction of (1). The full lines in Fig. 1, calculated from (1), show how good the agreement is. The energy gap, E_G , is plotted against temperature in Fig. 2.

From cyclotron resonance experiments⁴ we know that in germanium the energy maximum of the valence band is at the origin, and equal energy minima occur along the eight [111] axes of momentum in the conduction band. Therefore, in the above interpretation $k\theta_c$ is the energy of the longitudinal acoustic wave with momentum in the [111] direction equal to the momentum k_c of the electrons at the minimum of the conduction band. From the known values of the elastic constants⁵ and the theory of vibrations of the diamond lattice,⁶ we estimate that the conduction band minima occur at a momentum of magnitude $6.2 \times 10^7 \text{ cm}^{-1}$, which is about $\frac{2}{3}$ of the momentum at the edge of the zone in the [111] directions.

We have attempted to correlate the intrinsic carrier concentration n_i deduced by Morin and Maita⁷ from drift mobility and conductivity, with the values of E_G of Fig. 2. We find

$$n_i = 4.82 \times 10^{15} T^{\frac{3}{2}} (\bar{m}/m_0)^{\frac{3}{2}} N_c^{\frac{1}{2}} \exp(-E_G/2kT), \quad (2)$$

where $\bar{m} = 0.25m_0$ is calculated from the effective masses of holes and electrons⁴ including two valence bands and $N_c = 8$ is the number of minima in the conduction band. This gives, with $E_G = 0.655 \text{ eV}$, $n_i = 1.82 \times 10^{13} \text{ cm}^{-3}$ at 291°K, whereas Morin and Maita find $n_i = 1.4 \times 10^{13} \text{ cm}^{-3}$.

We have also estimated the magnitude of K theoretically. The factor A in (1) depends on the effective masses of holes and electrons, on k_c , θ , N_c , the refractive index, and the coupling constant C . This latter can be expressed in terms of the electron and hole mobilities. We find $A_{\text{theor}} = 1420 \text{ cm}^{-1}$, whereas the value of A which best fits the data is $A_{\text{obs}} = 1150 \text{ cm}^{-1}$. Although this close agreement may be fortuitous, we consider that

it does indicate that the magnitude predicted theoretically is of the right order.

Finally, we observe that using (2) with $\bar{m}=0.21m_0$, which brings the values of n_i of (2) and of Morin and Maita into line at 291°K, and values of n_i given by Morin and Maita at temperatures above 291°K, we have deduced E_G at higher temperatures. The results are shown dotted in Fig. 2, from which it will be seen that the two branches fit together with very little discontinuity of slope. We would remark on the quadratic behavior of E_G at low temperature.

Acknowledgment is made to the Chief Scientist, British Ministry of Supply, for permission to publish this letter.

¹ Hall, Bardeen, and Blatt, Phys. Rev. **95**, 559 (1954).

² H. Y. Fan and M. Becker, *Semi-conducting Materials* (Butterworths Publications, London, England, 1951).

³ H. B. Briggs, Phys. Rev. **77**, 287 (1950).

⁴ Lax, Zeiger, Dexter, and Rosenblum, Phys. Rev. **93**, 1418 (1954).

⁵ H. J. McSkimin, J. Appl. Phys. **24**, 993 (1953).

⁶ H. M. J. Smith, Trans. Roy. Soc. (London) **A241**, 105 (1948).

⁷ F. J. Morin and J. P. Maita, Phys. Rev. **94**, 1525 (1954).

Ultrasonic Attenuation in Metals by Electron Relaxation

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A DIFFERENCE in the ultrasonic attenuation between lead in the normal and superconducting states has been reported recently by Bömmel.¹ In this note it will be shown that the magnitude and temperature dependence of the attenuation in the normal state can be explained reasonably in terms of an incomplete adjustment of the Fermi distribution with respect to the elastic deformation, and consequently such an attenuation is to be expected in all metals at low temperatures when the mean free path becomes relatively long.²

In the free electron gas model of a metal, the Fermi surface is a sphere. With this gas we can associate an internal kinetic pressure given by $p_f = (2/5)nE_0$, where n is the number of electrons per unit volume and E_0 is the Fermi energy.³ If a longitudinal compressive strain ϵ_x (in the x -direction) is produced slowly, the Fermi surface remains spherical and p_f increases uniformly because the volume decreases. On the other hand, if ϵ_x is brought about quickly enough, only the electron velocity components in the x -direction react immediately and the Fermi surface is elongated momentarily in that direction. Collisions of electrons with the lattice eventually lead to the equilibrium spherical distribution, and the stress necessary to maintain ϵ_x relaxes to its equilibrium value. The relaxation time τ of this process is the same as the one commonly used in the theory of

electrical conductivity. The magnitude of the relaxing part of the stress Δp_x can be found simply by noting that the x -component of velocity is increased by a factor $(1+\epsilon_x)$, in the instantaneous application and by the factor $(1+\frac{1}{3}\epsilon_x)$ in the equilibrium case. In both situations n increases by $(1+\epsilon_x)$, and so Δp_x is found to be $(8/15)nE_0\epsilon_x \equiv b\epsilon_x$. A more detailed analysis also gives this result.

Such an effect may be expressed in terms of a relaxational elastic constant

$$k = k_0(1+b/k_0)[1+b/k_0(1+i\omega\tau)]^{-1}.$$

The attenuation constant α is the imaginary part of $-\omega(\rho_0/k)^{\frac{1}{2}}$, ρ_0 being the density. When $\omega\tau \ll 1$ we obtain, after expressing the relaxation time in terms of σ , the electrical conductivity, and neglecting b with respect to k_0 ,

$$\alpha = \frac{4}{15} \frac{\omega^2 m E_0 \sigma}{\rho_0 c_0^3 e^2}. \quad (1)$$

Here, c_0 is the longitudinal wave velocity, e is the electron charge, and m its mass.

Van den Berg⁴ observed that σ of lead in the normal state is given approximately by $1/\sigma = \rho' + 6.6 \times 10^{-13} T^5$ ohm-cm, where ρ' is the residual resistance. Assuming that curve R of Fig. 1 (which also shows Bömmel's

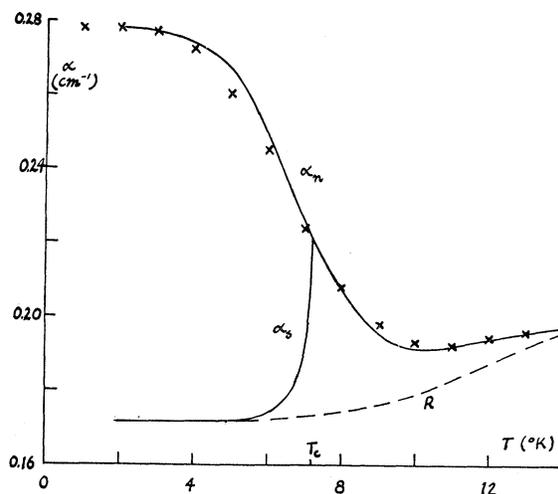


FIG. 1. Attenuation vs temperature in lead. The solid curves show Bömmel's measurements; curve R is assumed due to some other mechanism, and the crosses are calculated from Eq. (1).

results) is an attenuation due to some other cause, Eq. (1) can be used to evaluate ρ' by fitting $(\alpha_n - R)$ at the lowest temperature. Using $E_0 = 4$ eV and $c_0 = 2.4 \times 10^5$ cm/sec we obtain $\rho' = 1.0 \times 10^{-8}$ ohm-cm (a plausible magnitude). Attenuation at higher temperatures may be calculated using Van den Berg's results for σ and points are shown in Fig. 1.

The fair agreement between Eq. (1) and Bömmel's results in spite of the simple model used, lends support