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A STUDY OF RADIATION PRESSURE IN A REFRACTIVE MEDIUM BY THE PHOTON DRAG EFFECT

(Short Title: A Study of Radiation Pressure)

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ABSTRACT

The pressure of radiation in a refractive medium has been a matter of theoretical controversy for many years though relatively few experiments have been performed. We have measured the photon drag effect in germanium and silicon in the far infra red, up to a wavelength of 1.2 mm. At sufficiently long wavelengths the effect is independent of the semiconductor band structure and depends on the radiation pressure in the dielectric. We find that the expression originally deduced by Minkowski correctly describes our results.

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Introduction

"The momentum of a light wave in a refracting medium has been the subject of considerable controversy for more than half a century", (Burt and Peierls, 1973). Gordon (1973) quotes Blount as commenting that "the argument has not, it is true, been carried on at high volume, but the list of disputants is very distinguished". An outline of the controversy, with relevant references, has been given recently by Jones (1978). Up to 1973 the dispute was between the Minkowski (1910) formulation in which the radiation momentum is determined by $\underline{D} \times \underline{B}$ or the Abraham (1914) prediction that the momentum is proportional to $\underline{E} \times \underline{H}$. Burt and Peierls (1973), Peierls (1976) and others have argued that Abraham's result gives only the momentum residing in the electromagnetic field and pointed out that Abraham's tensor is symmetric and consistent with the relationship $E = mc^2$. On the other hand Minkowski's form is consistent with the de Broglie relation

$$p = h/\lambda = nh/\lambda_0$$

where p is the momentum of a photon in a medium of refractive index n and λ_0 is the free space wavelength of the light. Gordon (1973) and also Robinson (1975) argued that the difference arose because Minkowski's formulation included a mechanical component due to the reaction of the medium, though the latter interpretation appears to be disputed by Mikura (1976). In Gordon's view, supported by Peierls (1976), Minkowski's expression represents the pseudomomentum or "crystal momentum" whose conservation in an interaction is required by the translational symmetry of the medium surrounding the body involved in the interaction. Hence the Minkowski momentum determines the experimentally observed quantity in many situations (eg the radiation pressure on a reflecting surface at normal incidence) and Gordon (1973) has derived a theorem defining its

applicability. On this interpretation neither Abraham's nor Minkowski's formulations give the total radiation momentum which requires proper account to be taken of the modification of the field due to the polarisability of the atomic constituents of the refractive medium. This has been done by Peierls (1976, 1977) who has also considered experimental conditions in which Gordon's theorem would not apply and the effect of the atomic constituents observed. In summary the three derivations give the momentum, p , of a photon as

$$p(\text{Abraham}) = h/n\lambda_0 \quad (1)$$

$$p(\text{Minkowski}) = nh/\lambda_0 \quad (2)$$

$$p(\text{Peierls}) = \frac{h}{n\lambda_0} \left[\frac{1+n^2}{2} - \frac{q(n^2-1)^2}{2} \right] \quad (3)$$

where q is a measure of the field gradient at an atom compared with the macroscopic field and Peierls estimates $q \sim 0.2$. It may be noted that, for $q = 0$, Peierls' expression reduces to the arithmetic mean of the other two but for $q = 0.2$, p becomes negative for $n \geq 2.8$. We also note that for $n = 4$, $p(\text{Peierls}) = -p(\text{Minkowski})$ if $q = 0.218$.

Compared with the extent of the theoretical contributions on this topic, experimental work has been sparse. The Abraham force was observed in a suitably designed experiment by Walker and Lahoz (1975). Jones and his collaborators (Jones and Richards, 1954; Jones and Leslie, 1978) measured the impulse imparted to a mirror immersed in one of a series of liquids of differing refractive index and showed that the radiation pressure increased linearly with n , in agreement with the Minkowski expression (2). This result is to be expected at normal incidence on the basis

of Gordon's theorem but their results with polarised light at oblique incidence were not in agreement with Peierls 1977 prediction. Ashkin and Dziedzic (1973) observed that a focused laser beam, passing through an air-liquid interface in either direction, exerted a net outward force on the liquid. Qualitatively this result is consistent with both expressions (2) and (3) and covered by Gordon's theorem. Quantitative interpretation of this experiment is, however, complicated by the need to focus the laser to produce an observable liquid deflection: most theoretical work refers to "wide" beams (beam width greater than the light pulse duration multiplied by the velocity of sound) so that transverse forces are unimportant. This condition was not met by Ashkin and Dziedzic.

None of the above experiments measure photon momentum directly and indeed it is difficult to devise a practical experiment which does. In the circumstances it is clear that additional experimental information under any conditions not used hitherto would be valuable. The photon drag effect (Danishevskii et al, 1970; Gibson et al, 1970) is the generation of an electric field in a semiconductor due to the transfer of momentum from radiation to the electrons in the valence or conduction bands of the material. The momentum transfer process involved differs significantly from that operating in the reflection experiments of Jones et al and the high refractive index of convenient semiconductors (eg Germanium, $n = 4$) results in large numerical differences between the expressions (1), (2) and (3). On the other hand the magnitude and even sign of the photon drag field per unit intensity depend strongly on the characteristics of the semiconductor, particularly its electron energy band structure and electron scattering parameters. To obtain information on the photon momentum conditions must be chosen which minimise the effect of these parameters. We show in the next section that the necessary condition can

be met by making measurements at sufficiently long infra- red wavelengths.

The Photon Drag Effect

The photon drag effect is described by a fourth rank tensor but for the elemental semiconductors Ge and Si there are only two non zero, unequal tensor components, conventionally designated S and P (Cameron et al, 1975). The symmetric S component, observed when the wave vector of the light and the generated electric field are both collinear with a 100 crystal direction, has received most attention. Experimental results are frequently expressed in terms of S/σ , where σ is the absorption cross section. σ can be observed experimentally and varies relatively slowly with wavelength and temperature. The use of S/σ simplifies theoretical analysis since the absolute values of transition matrix elements are eliminated and, at least for strongly absorbing samples, it corresponds more nearly to the experimentally measured quantity.

In the analysis of photon drag data obtained to date (primarily in the wavelength range 2 to 12 μm and at room temperature) reliance has been placed on the requirement to conserve pseudomomentum in an optical transition (Gibson and Montasser, 1975). Experimental S/σ values in n-type Ge and Si are relatively low: this is ascribed to the requirement that a third particle (eg a phonon) co-operate in conduction band transitions to ensure simultaneous conservation of energy and pseudomomentum and some photon momentum is carried away by the co-operating particle (Gibson et al, 1970). In p-type material, on the other hand, direct intraband transitions are possible leading to large effects and a complex spectral response. Published measurements on p type germanium, using a variety of laser sources (Cameron et al, 1975; Gibson and Serafetinides, 1977; Al-Watban and Harrison, 1977) are collected together in Figure 1 together with the theoretically calculated

spectrum of Gibson and Montasser (1975). The latter, based on energy and pseudomomentum conservation together with the known band structure and phonon spectrum of germanium, adequately accounts for the main spectral features which include five sign reversals. The value of p , however, only appears in the absolute magnitude of S/σ . Though using the Minkowski value of p the fit shown is based on an argument that Gibson and Montasser admitted at the time was unconvincing so cannot be taken as evidence.

Information on photon momentum divorced from semiconductor effects can be obtained from photon drag measurements at light frequencies, ω , such that $\omega\tau < 1$ where τ is the electron mean free time. If $\omega\tau < 1$ the uncertainty in the electron energy, \hbar/τ , is large compared with the change in electron energy and similarly the uncertainty in electron pseudomomentum is larger than the pseudomomentum, \hbar/λ . The conservation of pseudomomentum and energy in a transition is then no longer required. Under these conditions the semiconductor band structure is irrelevant and the photon drag coefficient, S/σ , will reach a limiting, radiation pressure, value. Experimentally this condition will be characterised by S/σ becoming the same for n and p type material and independent of impurity concentration.

The electron scattering time, τ , is of course a function of electron energy and impurity concentration (see, for example, Smith 1979). The value of τ averaged over the electron (or hole) distribution function can however be estimated from a knowledge of the mobility and density of

states mass of the relevant charge carrier. At low impurity concentrations and room temperature we find τ , in picoseconds, to be 0.38 and 0.33 in n and p type germanium respectively and 0.2 and 0.12 in n and p type silicon. We therefore expect $\omega\tau \sim 1$ at a wavelength of about 700 μm in germanium and about 250 μm in p type silicon.

Photon Drag Measurements in the Far Infra Red

We have measured the photon drag coefficient, S/σ , of germanium and silicon samples at various wavelengths between 10 μm and 1.2 mm. The 100 orientated rod samples were prepared and measured much as in previous work (eg Cameron et al, 1975) except that relatively large (several mm) cross-section samples were required for measurements at the longest wavelengths. The large value of σ in both materials at long wavelengths necessitated the use of low impurity concentration, high resistivity material: about 20 ohm-cm germanium and 10 or greater ohm cm silicon. The former was still sufficiently removed from intrinsic to make corrections for the presence of the minority charge carrier small.

At wavelengths between 20 and 100 μm there is significant phonon absorption in germanium and silicon. We have allowed for the momentum lost directly to the lattice and the σ values refer only to absorption by charge carriers.

The radiation sources used were an H_2O vapour laser (28 and 33 μm), an HCN laser (337 μm) and various superfluorescent lasers optically pumped by appropriate lines of a CO_2 laser. The latter are tabulated in table 1. In all cases the pulse duration was $\lesssim 200$ ns so that the "wide beam" condition was met.

TABLE 1

<u>Lasing Molecules</u>	<u>CO₂ Pump Line</u>	<u>F.I.R. Wavelength</u>
D ₂ O	P32(9.63 μm)	66 μm
D ₂ O	R12(9.32 μm)	94 μm
D ₂ O	R12(9.32 μm)	114 μm
NH ₃	P32(10.72 μm)	151 μm
D ₂ O	R22(9.26 μm)	385 μm
CH ₃ F	P20(9.55 μm)	496 μm
¹³ CH ₃ F	P32(9.63 μm)	{ 1220 μm ~ 10% at 862 μm

It is worth noting that the development of CO₂ pumped far infra red sources - hundreds are now known eg Gallagher et al (1977) - has done more to open up the far infra red than any other development in recent years and has brought with it the need for new detector systems. The measurements reported here can be used to design photon drag devices with potentially valuable characteristics but we will describe these applications elsewhere.

The main problem in obtaining accurate results at long wavelengths is the measurement of laser power. We used a thermopile radiometer (Laser Instrumentation, Model 14BT) which had been constructed with a thicker coat of black paint than normal. The reflectivity of the thermopile was measured between 250 and 1250 μm (it increased from 2 to 46%) and what was not reflected was assumed to be absorbed. As the photon drag samples measured the pulse length the power of the laser lines could be obtained.

At wavelengths longer than 100 μm , where the signal to noise was very good, the accuracy of the results is estimated to be better than $\pm 10\%$.

Experimental Results

(i) Germanium

The results for n and p type germanium are shown in Figure 2. It will be seen that S/σ for n-type material rises monotonically from a small, positive, value at 10 μm to approach a limiting value at the longest wavelength measured (1.2 mm). P type germanium shows a positive peak near 0.03 eV, a sign reversal and a short negative region before becoming positive again and reaching the same limiting positive value attained by n-type germanium. In the notation of Gibson and Montasser (1975) the peak at 0.03 eV and the sign reversal is a "τ feature". It occurs when the initial energy of the electron in the light hole band, before a transition, equals the optic phonon energy. It is therefore the counterpart of the negative peak at 0.16 eV (Figure 1) which corresponds to the energy of the terminal state in the heavy hole band equalling the optic phonon energy. Neglecting band warping the ratio of the photon energies at the two peaks should therefore equal the effective mass ratio at low energies. Detailed computation (Montasser, private communication) including band warping predicts the peak at a photon energy of 0.031 eV as shown in Figure 2.

Most importantly, we note that Figure 2 shows the expected behaviour at long wavelengths, namely equal values of S/σ for both n and p material, though complete saturation is not achieved as $\omega\tau \sim 0.6$ at the longest wavelength measured.

(ii) Silicon

Experimental results for n and p type silicon are shown in Figure 3. Included in this figure are results at 33 μm and shorter wavelengths published by Serafetinides and Kimmitt (1978). The behaviour of n type silicon is similar to that of n type germanium since they have the same band structure. The broad, unresolved, positive peak between 0.02 and 0.04 eV in p-type silicon is due to two effects related to the band structure. Firstly a large positive peak is expected at 0.033 eV, which is the minimum separation of the heavy hole and split off bands (Kane 1956). At this point the bands are parallel and a peak occurs analogous to that in p type germanium at 0.26 eV and indicated in Figure 1 by an arrow (Gibson and Montasser, 1975). For photon energies less than 0.033 eV the only interband transitions allowed are between the heavy and light hole bands. These, however, run nearly parallel for separations greater than about 0.025 eV when the electron energy in the heavy hole band is only 0.02 eV or greater (Kane, 1956). Again, parallel bands lead to a large positive photon drag signal over a band centred around 0.025 eV. More important in the present context, however, is the behaviour at long wavelengths. As expected p type silicon saturates at a shorter wavelength than any of the three other materials due to its smaller value of τ_{and} becomes equal to n type material at the longest wavelength observed. We conclude that the condition $\omega\tau < 1$ is adequately, even if only marginally, met in all four materials at 1.2 mm and the limiting value of S/σ observed is independent of the band structure of the semiconductor under study.

Discussion and Conclusions

To obtain the theoretical limiting values of S/σ from the three expressions (1), (2) and (3) we simply multiply p by λ_0/c (Gibson and Montasser 1975) where c is the velocity of light in vacuum.

Numerical values may then be tabulated for comparison with experiment, as in Table 2.

TABLE 2

<u>S/σ (long wavelength limit) $\times 10^{-10} \text{ m}^{-1} \text{ A}^{-1}$</u>		
<u>$n = 4.0$, Germanium</u>	<u>$n = 3.4$, Silicon</u>	
+0.52	+0.61	Abraham
+8.33	+7.1	Minkowski
+4.4 ($q = 0$)	+3.85 ($q = 0$)	} Peierls
-7.3 ($q = 0.2$)	-3.0 ($q = 0.2$)	
-8.33 ($q = 0.218$)	-7.1 ($q = 0.32$)	

The experimental conditions conform to those considered in most theoretical treatments. In particular the duration of the illuminating laser pulse was sufficiently short to ensure "wide beam" conditions and sufficiently long compared with the relaxation time, τ , to ensure a steady state during almost the whole pulse duration. No reflection at an interface is involved and both materials show negligible dispersion throughout the wavelength range studied so no distinction between phase and group velocity is required.

There is the remote possibility of a sign ambiguity - sign reversals are not unfamiliar in photon drag experiments, as illustrated by p type germanium - but a positive photon drag signal is defined in the obvious

way, namely charge displacement in the same direction as the radiation wave vector.

Inspection of Figures 2 and 3 and comparison with the predicted numerical values in table 2 above shows immediately that the experimental value of S/σ at long wavelengths agrees with that predicted on the basis of the Minkowski expression for the photon momentum and differs from all other predictions by considerably more than the experimental uncertainty. We would not expect agreement with Peierls' expression since the electrons exchange momentum with the lattice and the experiment does not, by its nature, measure the total photon momentum which Peierls' theory describes. On the otherhand it is not obvious to us that Gordon's theorem on pseudomomentum conservation or Jones' (1978) argument are applicable under the conditions of our experiment so it would appear that Gordon's theorem is of wider applicability than considered hitherto. Central to Gordon's argument is the steady state condition which requires that any stress in the medium surrounding the interaction region be invariant in time. The mobile electrons and holes in a semiconductor can be considered as independent particles when present in low densities. They exchange momentum and energy with the surrounding medium by emission and absorption of phonons which equilibriate amongst themselves very rapidly, in a time much less than the laser pulse duration. Under these conditions Gordon's theorem might be expected to apply and experiment confirms that it does.

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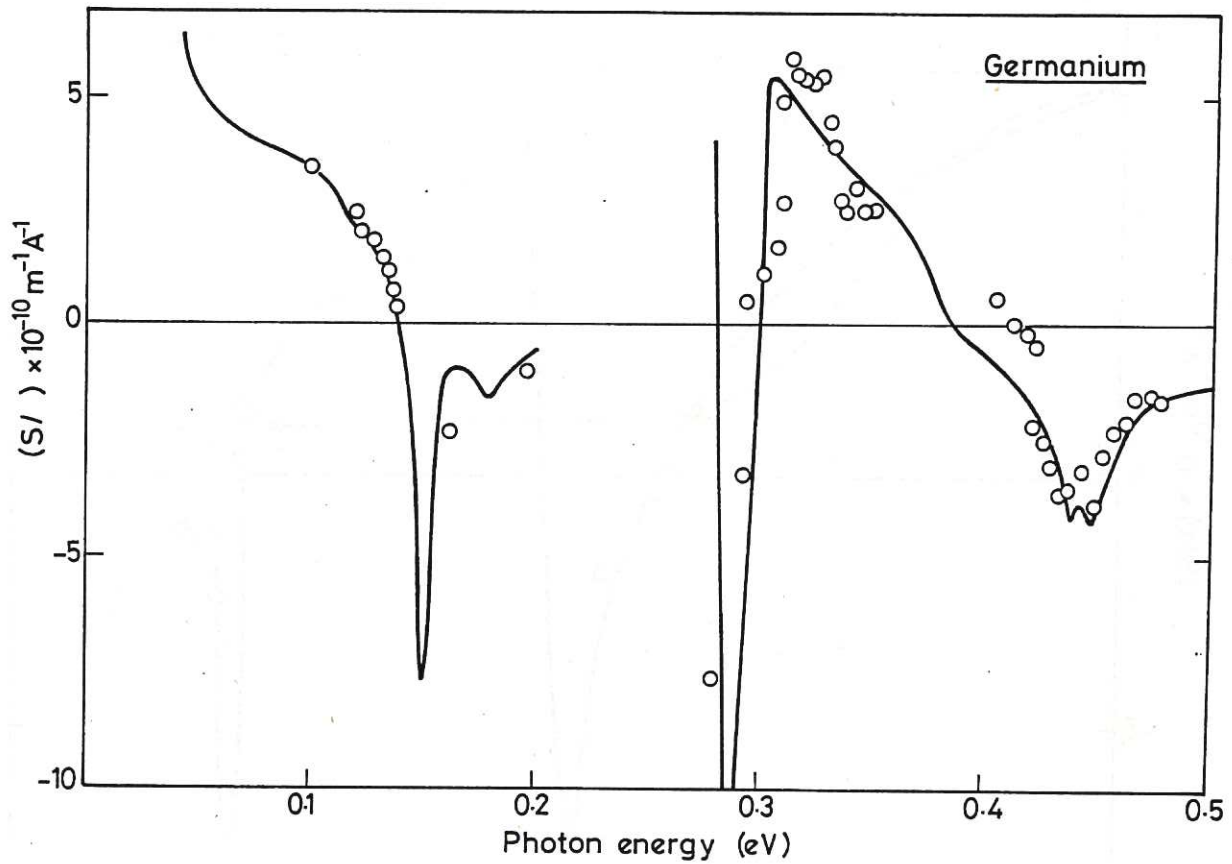


Fig.1 Collected experimental data on the photon drag coefficient, S/σ , of p-type germanium at room temperature. Curve is theoretical (Gibson and Montasser, 1975). The theory predicts an infinite positive peak near 0.27 eV but expansions used are then not valid.

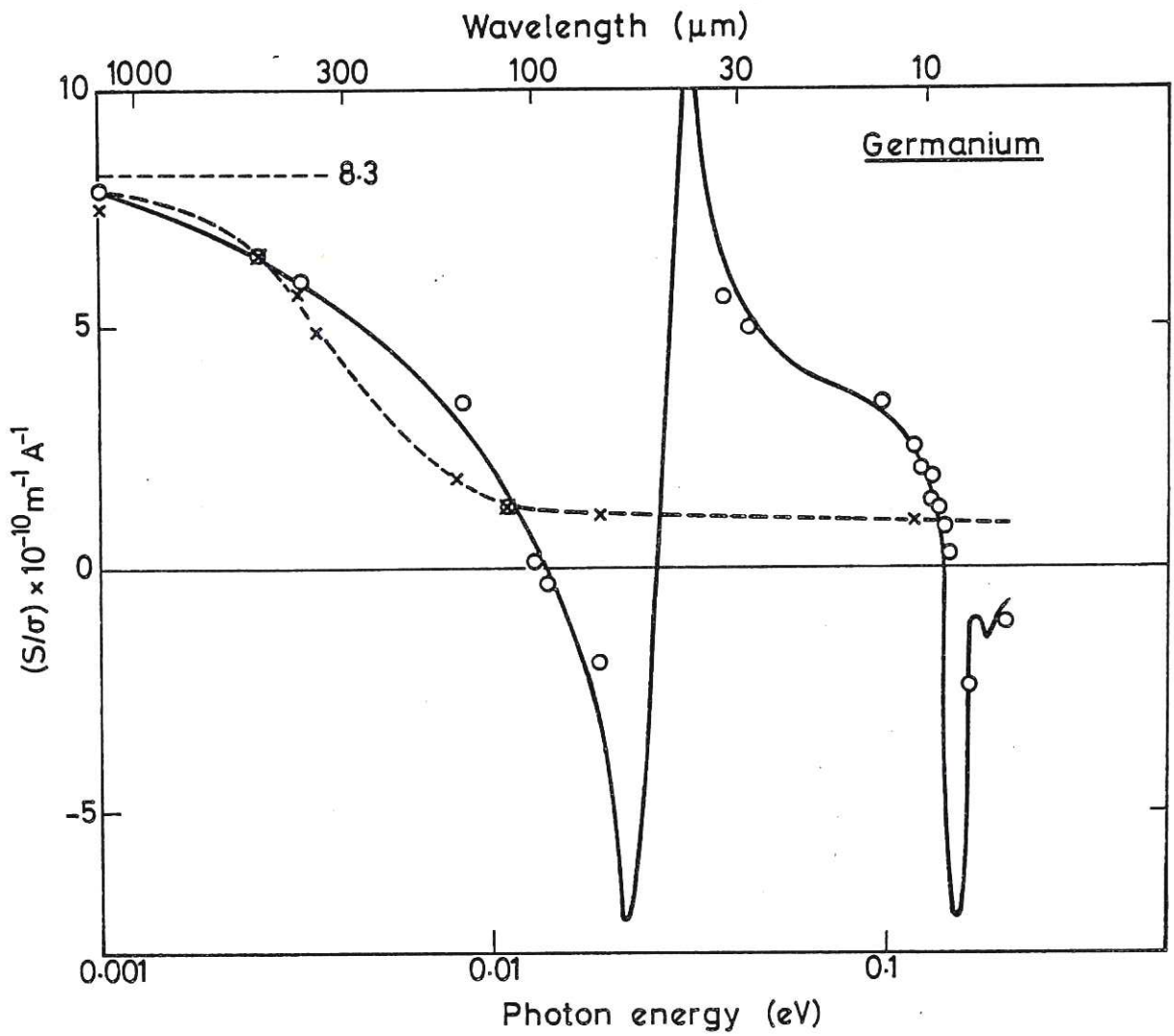


Fig.2 The photon drag coefficient, S/σ , of n and p type germanium in the far infra-red.
 —○— p type; —X— n type. On the basis of the Minkowski momentum tensor S/σ for both types should reach $8.3 \times 10^{10} \text{ m}^{-1} \text{ A}^{-1}$ at sufficiently long wavelengths.

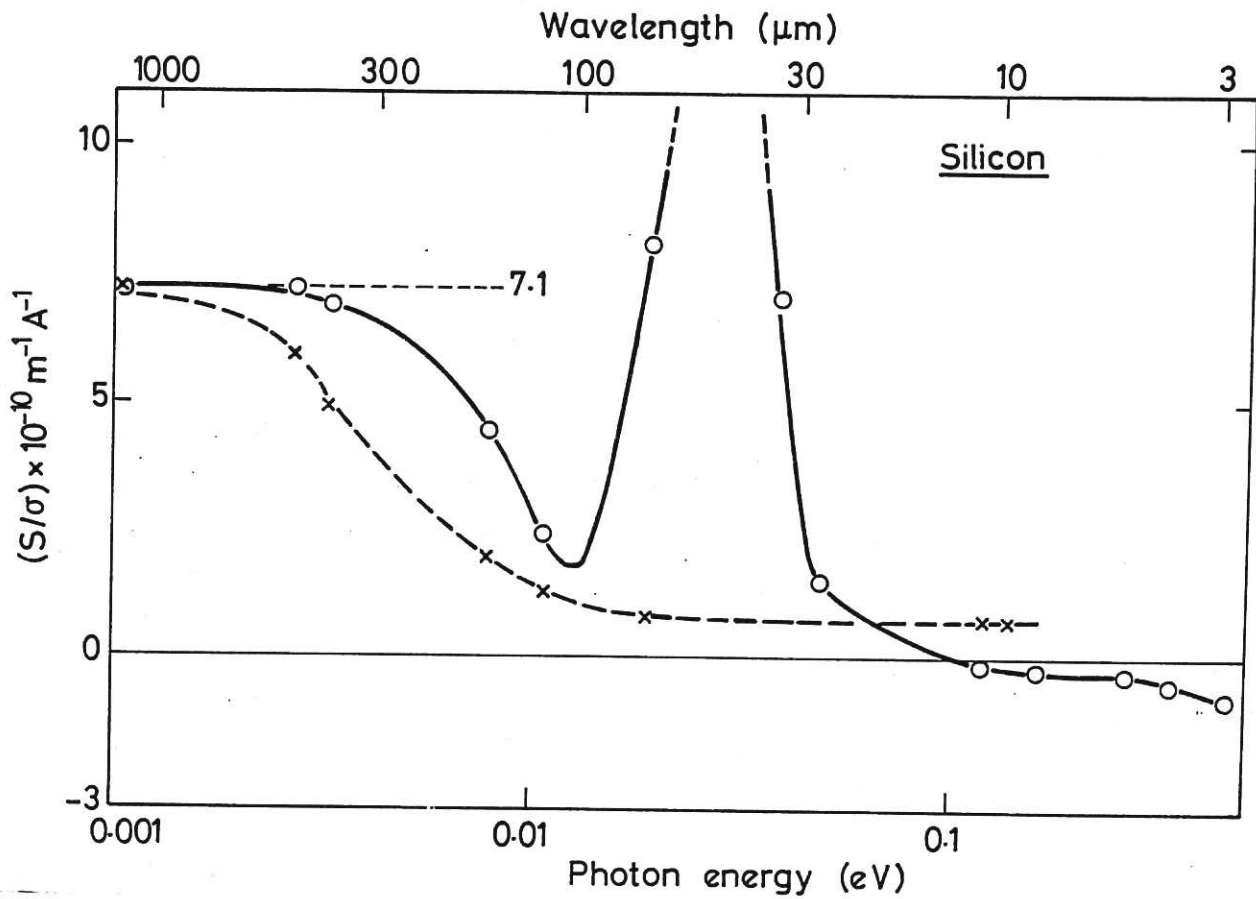


Fig.3 The photon drag coefficient, S/σ , of n type silicon in the far infra-red. On the basis of the Minkowski momentum tensor S/σ should reach $7.1 \times 10^{10} \text{ m}^{-1} \text{ A}^{-1}$ at sufficiently long wavelengths.



