PROCEEDINGS OF THE INTERNATIONAL CONFERENCE OF THE PHYSICS OF SEMICONDUCTORS, KYOTO, 1966
JOURNAL OF THE PHYSICAL SOCIETY OF JAPAN VOL. 21, SUPPLEMENT, 1966

VIII-1.

Scattering of Spin Polarized Carriers from Spin Polarized Impurities in Semiconductors*

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Scattering experiments with highly spin polarized (up to 98%) conduction electrons and highly spin polarized targets (neutral impurities) in phosphorus doped silicon have yielded the singlet and triplet scattering lengths, a_s and a_t, for the electron-neutral-phosphorus collision problem in the semiconductor. The ratio a_s/a_t is in approximate agreement with the calculated ratio for hydrogen, suggesting the applicability of the scaled hydrogen model for this problem. The high polarization is obtained through spin-lattice equilibrium in a magnetic field of 12 kOe and at a temperature of 0.35°K achieved with a liquid He^3 cryostat. Conduction electrons are produced at the low temperatures by impurity photoionization. In addition to making possible a detailed study of neutral impurity scattering, polarized spin photoconductivity experiments provide a new tool for investigating Zeeman spectroscopy and spin-lattice relaxation of impurities in semiconductors.

Semiconductors provide a convenient system on which to carry out scattering experiments where highly spin polarized "free" electrons scatter from highly polarized electron targets. The "free" electrons are those in the conduction band and the targets are neutral impurities. The high polarization is obtained as a consequence of the Boltzmann equilibrium at low temperature and high magnetic fields. The presence of electrons in the conduction band at the low temperatures can be assured by means of photoionization of the neutral impurities. The maximum polarization which can be attained for a $g\simeq 2$ electron spin system at a temperature of 0.35°K and magnetic field strength of 12 kOe is 98%, as can be determined from the relationship $P=\tanh g\mu_0 H/2kT$. These conditions for almost complete polarization are easily satisfied with a modest magnet and a liquid He^3 cryostat. Although the discussion in this paper is applicable to most semiconductors of either *p*- or *n*-type, we will confine ourselves to the specific case of *n*-type silicon containing phosphorus impurities at several concentrations.

The scattering results can be readily interpreted for hydrogenic-type impurities such as the group V donors in silicon or germanium, since the scaled hydrogen atom model can be expected to apply fairly accurately and since electron scattering from hydrogen has been subjected to extensive theoretical analysis. Indeed, neutral impurity scattering in silicon and germanium doped with shallow impurities has been treated as an electron-hydrogen atom collision analogue by Erginsoy[1)] and Sclar.[2)] The former used the hydrogen calculations of Massey and Moiseiwitsch[3)] and the latter took special account of a possibly bound singlet negative ion state analogous to the bound hydrogen negative ion. In neither of these works is special attention devoted to the properties of the scattering if the electrons and impurities are spin polarized, and it is this aspect with which the present paper is principally concerned. The investigation elucidates the nature of the impurity state and the appropriateness of the scaled hydrogen model, and also leads to an unambiguous determination of a neutral impurity scattering component even in the presence of other scattering mechanisms, since neutral scattering is the only type which is dependent on the spin polarization. This dependence arises because the antisymmetric requirement on the total wave function leads to different scattering amplitudes when the incident and scattering electron spins are parallel (triplet scattering) or antiparallel (singlet scattering) and because the degree of spin polarization determines the relative contributions of these two types of scattering.

The apparatus employed consists of a liquid He^3 cooled paramagnetic resonance spectrometer,

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* Research supported in part by the National Science Foundation and the Air Force Office of Scientific Research.

** Supported by a Xerox fellowship.

without the necessity of the microwave detection electronics. The operating temperature is between 0.35°K and 1.2°K and the microwave cavity in which the silicon sample is placed is tuned near 25 Gc/s. A light pipe with a blackened sliding plunger introduces black body radiation onto a sample equipped with alloyed leads for photocurrent measurements. The black body plunger can assume any temperature between 1°K and 290°K by virtue of its position in the light pipe and its temperature is determined by means of an imbedded thermocouple. A CaF_2 reflector adjacent to the sample has also been used to obtain selective reflection of photons of $\lambda > 20\ \mu$ using the room temperature black body radiation from the top of the light pipe as the source. Three phosphorus doped samples are reported on in this paper. They are uncompensated and contain phosphorus densities of $1.8 \times 10^{17}/\text{cm}^3$, $3.6 \times 10^{16}/\text{cm}^3$ and $4.5 \times 10^{14}/\text{cm}^3$.

For low energy incident electrons, the scattering cross-section of a hydrogen atom can be written in terms of the scattering lengths a_s and a_t, corresponding respectively to singlet and triplet type scattering. One gets

$$\sigma_{\text{neutral}} = s(4\,\pi a_s^2) + t(4\,\pi a_t^2)\ . \qquad (1)$$

s and t are respectively the fractions of singlet and triplet type collisions. For an unpolarized beam or target, $s=1/4$ and $t=3/4$. For a completely polarized beam and target, $s=0$ and $t=1$. The singlet scattering contribution is a maximum for an unpolarized beam and goes monotonically toward zero as the polarization increases. This is important because $a_s > a_t$ for hydrogen, and thus the total neutral scattering cross-section is expected to decrease and mobility is expected to increase as the polarization P increases. This is indeed found to be the case in our investigations.

The problem can be treated quantitatively by computing s and t as a function of P, assuming

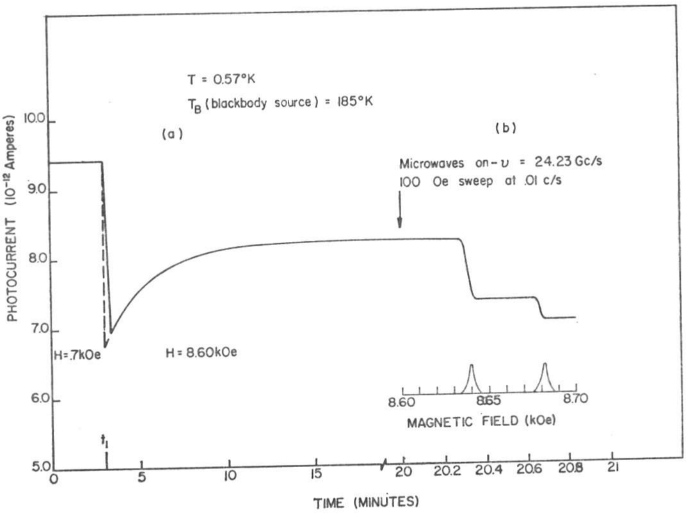

Fig. 1. Two methods for obtaining $I_H(P)$ at different values of spin polarization. Sample contains $3.6 \times 10^{16}/\text{cm}^3$ phosphorus impurity density. $\boldsymbol{I} \| [100]$ direction. $\boldsymbol{H}$ is along [110] direction making 45° angle with $\boldsymbol{I}$. (a) is applicable when the spin-lattice relaxation time T_1 is long. At t_1, H is switched from 0.70 kOe to 8.6 kOe. The sudden current drop at t_1 is due to magnetoresistance and the subsequent current rise is due to the increase of polarization through the spin-lattice relaxation. $I_H(0)$ at $H=8.6$ kOe is obtained from the dashed line extrapolation at t_1 for instantaneous change of magnetic field. $I_H(P_0)$ at 8.6 kOe is the equilibrium value of the current corresponding to the equilibrium polarization P_0. (b) Starting with $I_H(P_0)$ at 8.6 kOe, the low field resonance associated with the hyperfine structure of phosphorus is first saturated, changing the current to $I_H(P_0/2)$. Then the low field resonance is saturated, changing the current to $I_H(\sim 0)$. Due to spin exchange effects, there remains a small polarization which can be reduced to zero for this sample by traversing the resonance several times.

as is almost always the case, that impurity and free electrons have the same P. One obtains $s=(1-P^2)/4$ and $t=(3+P^2)/4$. Upon substituting these values into eq. (1), we find that the dependence of σ_{neutral} on P is given by

$$\sigma_{\text{neutral}}=\pi[(a_s^2+3\,a_t^2)-(a_s^2-a_t^2)P^2]\,. \quad (2)$$

If other scattering mechanisms are present, such as ionized impurity or defect scattering, the sum of their scattering cross-sections (weighted by their relative number), σ_0, can be added to σ_{neutral} in order to obtain a total cross-section. Since the reciprocal of the photocurrent, I, is proportional to the total cross-section, we obtain

$$1/I=K(H)[(a_s^2+3\,a_t^2)-(a_s^2-a_t^2)P^2+\sigma_0]\,, \quad (3)$$

where $K(H)$ depends on the magnetic field H through the magnetoresistance and is otherwise dependent only on the concentration, the applied electric field and the sample geometry. To determine the ratio a_s/a_t, it is necessary to know $\sigma_0/\sigma_{\text{neutral}}$ and to know, for two different polarizations, $I_H(P)$, which is the value of I at fixed H. $\sigma_0/\sigma_{\text{neutral}}$ can be obtained through mobility measurements on samples of various concentrations, complemented by lifetime and other transport determinations, and $I_H(P)$ can be obtained by either of two methods, both of which are shown in Fig. 1. The first method is applicable when the spin-lattice relaxation time, T_1, of the impurities is long compared with the time required to change the magnetic field by a substantial fraction of its maximum strength. Starting at a low value of the magnetic field where P is substantially zero, the field is raised to a high value H (8.6 kOe in Fig. 1a). Since the time to increase the magnetic field is much less than T_1, the initial current at 8.6 kOe corresponds to an almost unpolarized state, showing a reduced value because of magnetoresistance. Subsequently, due to the spin-lattice relaxation, the polarization P increases until it reaches its equilibrium value $P_0(H)$, and this is accompanied by a rise in photocurrent. Thus, by an extrapolation to a zero switching time of the magnetic field, which is indicated in the figure by the dashed line, one obtains $I_H(0)$, and from the equilibrium value of the current, $I_H(P_0)$ is determined; these two values provide the requirements of our experiment. The second method is more generally applicable, in that it does not depend on the value of T_1. $I_H(P_0)$ is measured at a magnetic field value in the vicinity of the impurity Zeeman resonances corresponding to an available microwave frequency. Then the resonance is traversed with saturating microwave power, and the decrease in photocurrent follows from the reduced spin polarization. This is seen in Fig. 1b, where the resonance fields of approximately 8.64 kOe and 8.68 kOe correspond to the microwave frequency of 24.23 Gc/s for $g\simeq 2$. The two resonances arise because of the hyperfine structure associated with the spin 1/2 value of the phosphorus impurity nucleus. Saturation of the first line reduces P_0 to $P_0/2$, giving a current after the resonance traversal denoted by $I_H(P_0/2)$. For the particular sample involved in Fig. 1, T_1 happens to be sufficiently long so that $I_H(\sim 0)$ can also be obtained simply by saturating the second resonance within a short time($\ll T_1$) after the first resonance, making the saturation cumulative except for small spin exchange[4] effects. Upon repeated saturation of both lines, $I_H(0)$ can be obtained.

By employing the above methods for various H and T values, it is possible to produce a plot of essentially $1/I$ against P^2, which is shown in Fig. 2. The linear dependence is in agreement with eq. (3), and the slope of the experimental curve yields an a_s/a_t ratio of 2.0, assuming $\sigma_0 \ll \sigma_{\text{neutral}}$. The solid line is a theoretical plot based upon calculated values[5] of a_s and a_t for hydrogen which gives an a_s/a_t ratio of 2.65.

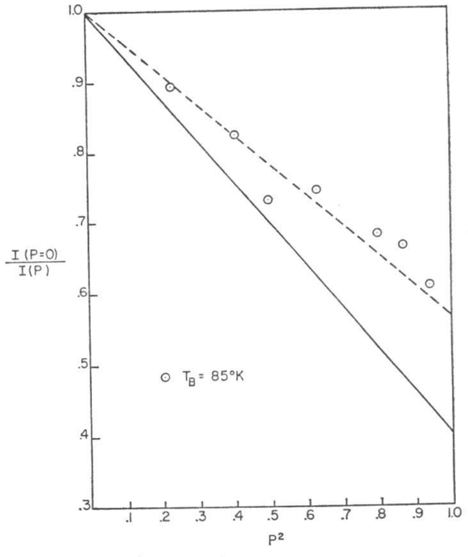

Fig. 2. Dependence of $1/I$ on P^2. Linear dependence is in accord with eq. (3). Solid line is theoretical curve based on eq. (3) and on calculated atomic hydrogen scattering lengths. Silicon sample is same as that of Fig. 1.

The discrepancy is not large, but it is worth inquiring into its cause.

When T_B of 290°K source temperature is used, a linear dependence between $1/I$ and P^2 still results but the slope has only about half the magnitude of that corresponding to the 85°K source. It thus represents a stronger departure from the theory. With a $2\,\mu$ light source, the dependence on polarization practically disappears. These results are not surprising, since for conduction electron kinetic energies exceeding about 1/4 the impurity ionization energy, the scattering probabilities should be energy dependent. From these differences encountered with sources of different temperatures, it can be concluded that the photoexcited electrons do not thermalize before capture; *i.e.* the velocity distribution differences are maintained throughout the carriers' lifetimes. For the $3.6\times10^{16}/\mathrm{cm}^3$ sample referred to in the two figures, the compensation is very low as determined from lifetime measurements.6) The lifetime exceeds 10^{-9} seconds and the thermalization time must be longer than that. Now even with the 85°K source, whose peak in the black body radiation spectrum is at $33\,\mu$, a significant percentage of the ionized carriers have initial kinetic energy exceeding 1/4 of the impurity ionization energy. It is thus possible that the discrepancy in a_s/a_t is due to this energy difference. Other possibilities include an inadequacy of the scaled model because of displacement of the bound or near-bound singlet negative phosphorus ion state with respect to the scaled hydrogen negative ion state, or the presence of a σ_0 which is not entirely negligible compared with $\sigma_{\mathrm{neutral}}$.

Preliminary transport and lifetime measurements have been made on the various samples. For the $3.6\times10^{16}/\mathrm{cm}^3$ sample from which the preceding results were obtained, the lifetime measurements indicated that the compensating acceptor concentration is at least two orders of magnitude smaller than the phosphorus concentration. Thus it is probable that the ionized impurity scattering probability is small in comparison with the neutral impurity scattering probability. The mobility of electrons in this sample, photoionized by light with $\lambda>20\,\mu$ (300°K black body radiation reflected from $\mathrm{CaF_2}$), is $5.8\times10^3\,\mathrm{cm^2/V\,sec}$ at 1.1°K, which is about a factor of 3 lower than μ_{neutral} predicted by Sclar at this temperature; however, his theory has not been subjected to experimental test at these low temperatures. Mobility measurements on a $1.8\times10^{17}/\mathrm{cm}^3$ sample were hampered by the very large absorption coefficient of light of $\lambda>20\,\mu$ and the attendant inhomogeneous photoelectron concentration in our relatively thick sample (1 mm). A value near $2\times10^3\mathrm{cm^2/V\,sec}$ was obtained but it is not very reliable. The a_s/a_t ratio measured from polarized carrier experiments on this $1.8\times10^{17}/\mathrm{cm}^3$ sample corresponded closely to that obtained for the $3.6\times10^{16}/\mathrm{cm}^3$ sample. An additional effect which could come into play in the concentrated sample is interferences of scattering amplitudes among close neighbors. A dilute sample of $4.5\times10^{14}/\mathrm{cm}^3$ had a measured mobility of $6.0\times10^4\,\mathrm{cm^2/V\,sec}$. It is seen from the upper bound on neutral impurity scattering probability for the $3.6\times10^{16}/\mathrm{cm}^3$ sample and from the relative mobilities that, at most, neutral impurity scattering contributes 15% to the total scattering probability in this sample. Thus, we might expect a 7% change in current between $P=0$ and $P=1$ for the most favorable T_B of 85°K. We have looked for this, but have been unable thus far to detect any polarization dependence.*** It is possible that another effect is responsible for quelling the polarization dependence in dilute samples. The carriers can drift across the sample before spending enough time on the impurities to become polarized. This would depend on carrier lifetime, drift velocities, velocities under the applied field, impurity concentration, rate of ionization and T_1 on the impurities. Because the T_1, even under illumination, is very long in silicon for dilute samples, we estimate this effect could become important at concentrations in the $10^{14}/\mathrm{cm}^3$ range.

In conclusion, a method using highly polarized carriers for studying neutral impurity scattering in semiconductors has been described, with experimental application to silicon. The technique also serves as a new means for measurement of spin-lattice relaxation, Zeeman spectroscopy of impurities, and velocity distribution of photoexcited carriers at low temperatures.

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*** Note added in proof: The expected current change with polarization has since been observed.

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DISCUSSION

Hasegawa, H.: You have mentioned that you got different results of apparent polarization degrees of the cross-section for different temperatures of the black body radiation. Does this mean that the polarization of the produced carriers would be dependent on the temperature of the black body radiation?

Honig, A.: The spin polarization of the photo-ionized carriers should depend very little, if at all, on the temperature of the black body radiation source, for the range of radiation temperature we used. This is concluded from independent magnetic resonance experiments, where no change in electron spin temperature was observed under illumination. The reason for the change of the $(I)^{-1}$ vs. P^2 curve is probably the dependence of the singlet and triplet scattering cross-sections on carrier velocity, which depends on the black body source temperature.