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# Shubnikov–de Haas effect study of InAs after transmutation doping at low temperatures

H Gerstenberg and P Müller†
Fakultät für Physik E21, Technische Universität München, Reaktorstation, D-8046 Garching, Federal Republic of Germany

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**Abstract.** Degenerate InAs single crystals have been irradiated by thermal neutrons below 6 K. The Shubnikov–de Haas effect and the electrical resistivity have been measured as a function of the neutron dose and the annealing temperature. The effects of transmutation doping and simultaneous introduction of lattice defects have been analysed in terms of the conduction electron density and the scattering rates $\tau_\rho^{-1} = \rho n e^2/m^*$ and $\tau_x^{-1} = 2\pi k_B X/\hbar$ (where $X$ is the Dingle temperature). The measured conduction electron density after irradiation and thermal annealing agreed well with the values calculated from the experimental and materials parameters. The effects of radiation damage may qualitatively be explained assuming neutral In vacancies to be the most common type of defect in thermal-neutron-irradiated InAs. A comparison with similar experiments on InSb is given.

## 1. Introduction

Neutron transmutation doping is a useful preparation technique for semiconductors which provides high homogeneity and adjustability of the doping profile. However, the method has achieved technological importance only in the case of Si:P, whereas its application to compound semiconductors is rather infrequent. In particular, to the best of our knowledge, only one paper on the transmutation doping of InAs has been published so far [1].

The irradiation of InAs with thermal neutrons leads to the introduction of Sn and Se impurity atoms due to nuclear reactions in a concentration given by

$$n_{\mathrm{NTD}} = \sum_i (n_i \sigma_i) \Phi_{\mathrm{th}} t_{\mathrm{irr}} \tag{1}$$

where $\Phi_{\mathrm{th}}$ is the flux of thermal neutrons and $t_{\mathrm{irr}}$ the time of exposure. The product $(n\sigma)$ of the particle density and the neutron capture cross section determines the transmutation rate for each isotope contained in the sample. In the case of InAs the sum has to be taken over the three isotopes $^{113}$In ($n\sigma = 8.8 \times 10^{-3}\ \mathrm{cm}^{-1}$), $^{115}$In ($n\sigma = 3.48\ \mathrm{cm}^{-1}$) and $^{75}$As ($n\sigma = 0.077\ \mathrm{cm}^{-1}$). From these values it can be seen that the transmutation doping of InAs is almost entirely due to the reaction

$$^{115}\mathrm{In(n, }\gamma)^{116}\mathrm{In} \xrightarrow{\beta} {}^{116}\mathrm{Sn} \tag{2}$$

which takes place via two reaction channels with half-lives $T_{1/2}$ of 14.2 s and 54.2 min,

† Present address: Walther-Meißner-Institut, Walther Meißner Straße, D-8046 Garching, Federal Republic of Germany.

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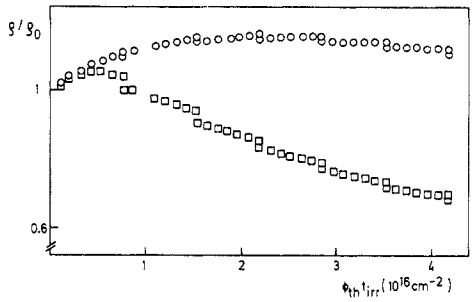


**Figure 1.** Resistivity $\rho(T = 4.6\,\mathrm{K})$ of two InAs crystals as a function of the thermal neutron dose $\Phi_{th}t_{irr}$: ○, $n_0 = 2.0 \times 10^{17}\,\mathrm{cm}^{-3}$; □, $6.8 \times 10^{16}\,\mathrm{cm}^{-3}$. The resistivities have been normalised to the values of the unirradiated samples.

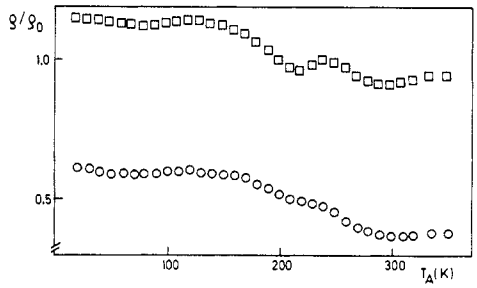


**Figure 2.** Resistivity $\rho(T = 4.2\,\mathrm{K})$ of InAs as a function of the annealing temperature $T_A$: □, $2.0 \times 10^{17}\,\mathrm{cm}^{-3}$; ○, $6.8 \times 10^{16}\,\mathrm{cm}^{-3}$. The duration of each annealing step was 5 min. The resistivities have been normalised to the values of the unirradiated samples.

respectively. In InAs all transmutation products are known to be donors when placed on the regular lattice positions of the corresponding mother isotope.

In addition to the nuclear reactions the neutron irradiation of InAs leads to the introduction of lattice defects. The mean recoil energy transferred to a $^{116}$In atom in the (n, $\gamma$) process can be estimated to be about 50 eV [2], which exceeds the displacement energies $E_d$(In) = 6.5 eV and $E_d$(As) = 8.5 eV [3] of InAs by far. However, only isolated point defects are created in this way, whereas a correlated defect structure is only produced by the irradiation with fast neutrons, which form a part of any reactor spectrum. Generally both kinds of defect can be electrically charged and thermally unstable.

The Shubnikov–de Haas (SdH) effect has proved to be a suitable technique for measuring the electronic properties of degenerate semiconductors. In narrow-gap materials such as InAs possessing a spherical Fermi surface the charge carrier density $n$ and the SdH frequency $F$ are related by the simple equation

$$n = 1/3\pi^2(2e/\hbar)^{3/2}F^{3/2}. \tag{3}$$

Moreover the Dingle temperature $X$ describing the attenuation of the SdH amplitudes is proportional to the scattering rate of the conduction electrons.

## 2. Experiments

The starting material for the irradiation experiments were degenerate single-crystal n-type InAs wafers with charge carrier densities $n_0$ between $5 \times 10^{16}$ and $2.0 \times 10^{17}\,\mathrm{cm}^{-3}$ at 4.2 K, which had been cut into rectangular parallelepipeds of approximately 2 mm × 5 mm. The maximum sample thickness was 0.5 mm in order to avoid self-shielding during the irradiation.

All experiments have been performed at the low-temperature irradiation facility of the Munich Research Reactor [4]. The sample temperature during irradiation was below 6 K. The flux of thermal and fast ($E > 0.1$ MeV) neutrons in the irradiation position were $\Phi_{th} = (1.1 \pm 0.1) \times 10^{13}\,\mathrm{cm}^{-2}\,\mathrm{s}^{-1}$ and $\Phi_f = (1.1 \pm 0.1) \times 10^{12}\,\mathrm{cm}^{-2}\,\mathrm{s}^{-1}$, respectively.

The experimental procedure started with the measurement of the SdH effect of the unirradiated specimens. During the subsequent low-temperature irradiation the

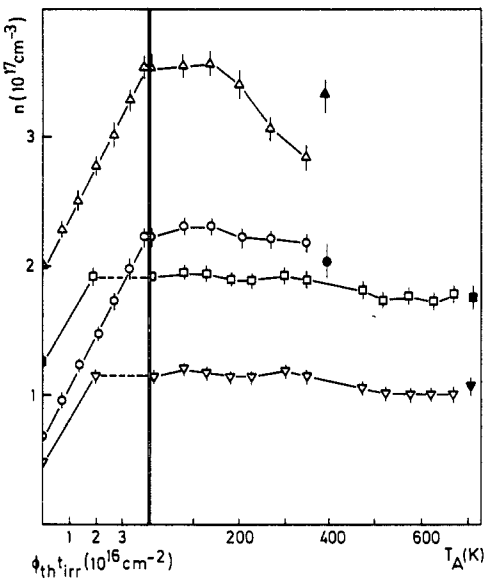


**Figure 3.** Conduction electron density $n$ of four InAs samples as a function of the thermal neutron dose $\Phi_{th}t_{irr}$ and the annealing temperature $T_A$. The sums of initial and calculated transmutation doping are given as full symbols. The reason for the decrease in $n$ by annealing the sample with the highest carrier density could not be resolved.

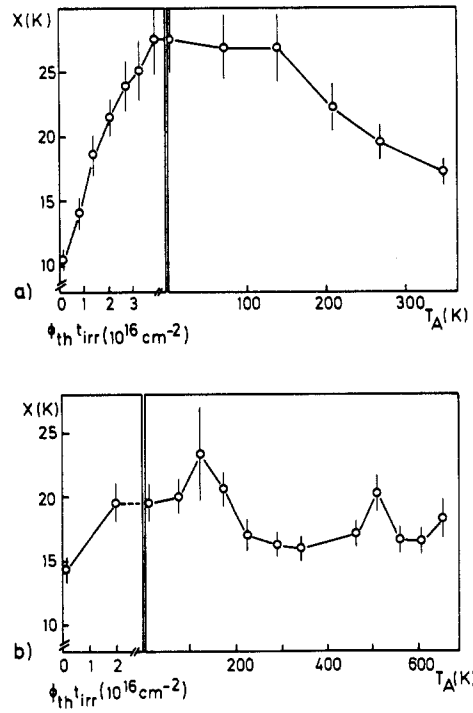


**Figure 4.** Dingle temperature $X$ of InAs as a function of the thermal neutron dose $\Phi_{th}t_{irr}$ and the annealing temperature $T_A$: (*a*) $n_0 = 6.8 \times 10^{16}\,cm^{-3}$; (*b*) $n_0 = 1.3 \times 10^{17}\,cm^{-3}$.

electrical resistivity of the samples was monitored as a function of the thermal neutron dose. After a decay time of at least 10 h, i.e. a long time compared with the half-life of $^{116}$In, the SdH effect of the irradiated samples was measured without intermediate warming up. Maximally six consecutive irradiations up to an integrated dose $\Phi_{th}t_{irr}$ of $4 \times 10^{16}\,cm^{-2}$ were performed. Subsequent to the irradiations an isochronal thermal annealing programme in the temperature range between 20 and 350 K has been carried out. Annealing experiments between $T_A = 450$ K and $T_A = 650$ K were performed after the final cooling of the radioactivity of the samples about 4 weeks after the irradiation. In all cases the measuring temperature was 4.2 K. A detailed description of the experimental set-up and the data analysis has been published previously [5].

## 3. Results

Figure 1 shows the resistivity $\rho(T = 4.6\,K)$ of two InAs samples with different initial carrier densities $n_0$ as a function of the thermal neutron dose. For an easier comparison between the two curves the resistivities have been normalised to the values of the unirradiated samples. Their absolute values are $\rho(T = 4.6\,K) = 0.8\,m\Omega\,cm$ for $n_0 = 2 \times 10^{17}\,cm^{-3}$ and $\rho(T = 4.6\,K) = 1.8\,m\Omega\,cm$ for $n_0 = 6.8 \times 10^{16}\,cm^{-3}$. An irradiation-induced increase in the resistivity was found only in the region of relatively small neutron

fluences whereas the resistivity decreased in spite of the increasing neutron dose for longer times of exposure. The most significant changes in the $\rho = f(\Phi_{th} t_{irr})$ characteristic have been observed in the sample with the lowest $n_0$. The neutron irradiations have been divided into up to six single steps (see above) in order to measure the SdH effect as a function of the neutron dose. Subsequent to each irradiation step—corresponding to a time of exposure of typically 10 min—the irradiation was therefore interrupted for several hours. During this time the resistivity of all samples further decreased to its final value, obeying an exponential law in time:

$$\rho(t) - \rho(t = \infty) = [\rho(t = 0) - \rho(t = \infty)] \exp\{[(-\ln 2)\, t]/T_{1/2}\}. \tag{4}$$

The time constant $T_{1/2}$ of this decay equals the half-life of $^{116}$In, indicating the introduction of additional donor-type impurities by $\beta$-decay. This post-irradiation decrease in the resistivity leads to clearly visible steps in the $\rho$ versus $\Phi_{th} t_{irr}$ curves in figure 1.

The effect of thermal annealing on the resistivity is shown in figure 2. The data, which again have been normalised to the values of the starting materials, correspond to the $\rho = f(\Phi_{th} t_{irr})$ curves in figure 1.

Figure 3 shows the change in the conduction electron density of InAs as determined from the SdH frequencies according to equation (3) as a function of the thermal neutron dose and the annealing temperature. Up to an integrated dose of $\Phi_{th} t_{irr} = 4 \times 10^{16}$ cm$^{-2}$ an almost linear increase in carrier density was found, showing an electron introduction rate $\Delta n/(\Delta \Phi_{th}\, t_{irr})$ of $4.05 \pm 0.61$ cm$^{-1}$. This value is independent of the initial doping level; it exceeds the pure transmutation rate (as calculated by equation (1)) by about 12%. No major changes in the carrier density have been introduced by thermal annealing. After heating the samples to 670 K the measured conduction electron densities equalled, to within the error bars, the sum of initial and transmutation doping as is shown in figure 3 by full symbols.

The Dingle temperatures $X$ of the InAs samples have been determined by fitting the SdH oscillation profiles at fixed measuring temperature [5]. For unirradiated specimens the $X$-values were found to increase with increasing carrier density exhibiting values between $10.4 \pm 0.5$ K ($n_0 = 6.8 \times 10^{16}$ cm$^{-3}$) and $14.3 \pm 0.9$ K ($n_0 = 1.3 \times 10^{17}$ cm$^{-3}$). These results are in good agreement with the data published so far [6–8]. The change in the Dingle temperature by thermal neutron irradiation and subsequent annealing is shown in figure 4. The irradiation-induced increase in $X$ is mainly annealed between $T_A = 100$ K and $T_A = 300$ K. However, even after the annealing programme had been completed, the samples show significantly higher $X$-values than those of the starting material. This observation was to be expected because of the increased impurity content introduced by neutron transmutation.

## 4. Discussion

In the experiments described above, the neutron transmutation doping leads to an irreversible change in the conduction electron density and the electrical resistivity. The irradiation damage and the annealing behaviour of InAs will therefore be discussed in terms of the scattering rates

$$\tau_\rho^{-1} = \rho n e^2/m^* \tag{5}$$

and

$$\tau_X^{-1} = 2\pi k_B X/\hbar. \tag{6}$$

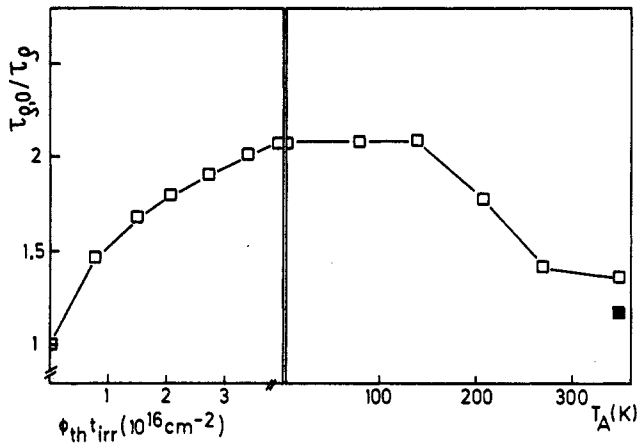


**Figure 5.** Transport scattering rate $\tau_\rho^{-1}$ of InAs ($n_0 = 6.8 \times 10^{16}$ cm$^{-3}$), as normalised to the value $\tau_{\rho 0}^{-1}$ of the unirradiated sample versus thermal neutron dose $\Phi_{th}t_{irr}$ and annealing temperature $T_A$, The increased $\tau_\rho^{-1}$ due to ionised impurity scattering by the transmutation-enhanced dopant concentration is given as full square. This value has been evaluated using equation (7).

It is well known [9] that these two scattering rates are not equivalent, because $\tau_X^{-1}$ is determined by large- and small-angle scattering whereas $\tau_\rho^{-1}$ is sensitive to large-angle scattering events only.

Figure 5 shows the scattering rates $\tau_\rho^{-1}$ of InAs ($n_0 = 6.8 \times 10^{16}$ cm$^{-3}$) as a function of the thermal neutron dose and the annealing temperature. The $\tau_\rho^{-1}$-values have been deduced using the measured $\rho$ and $n$ data; the effective masses $m^*$ have been evaluated from $n$ by means of the two-band model assuming that $m_0^* = 0.023m_e$ where $m_e$ is the free-electron mass [10]. The resistivity of degenerate semiconductors at low temperatures is mainly determined by screened ionised impurities. Using the Born approximation the corresponding scattering rate can theoretically be described by (see, e.g., [11])

$$\frac{1}{\tau_{\rho,ii}} = \left(\sum_i (N_i Z_i^2) e^4 m^* \Big/ 24\pi^3 (\varepsilon_0 \varepsilon_s)^2 \hbar^3 n\right)\left(\ln(1+b) - \frac{b}{1+b}\right) \qquad (7)$$

$$b = 4k_F^2 R_{TF}^2$$

where $N_i$ is the concentration and $Z_i e$ the charge of the ionised impurities contained in the sample; $n$ is the conduction electron density, $k_F$ is the Fermi wavenumber, $R_{TF}$ is the Thomas–Fermi screening length and $\varepsilon_s = 14.55$ [12] is the static dielectric constant of InAs. For all our samples a $b$-value of at least 10 has been evaluated, fulfilling the criterion $b \gg 1$ for the applicability of the Born approximation [13].

Although a large variety of different defects is possible in compound semiconductors, it is reasonable to think of singly ionised donor ions as the most important scattering centres in unirradiated or completely annealed samples. In this case the $n$-dependence of the scattering rate (7) is determined by the increase in the effective mass and the term in parentheses with increasing carrier density only. Taking into account this effect, the scattering rate corresponding to the enhanced doping level after neutron transmutation has been calculated by means of equation (7) and inserted in figure 5 as a full square. It can be seen from the figure that evident annealing of InAs takes place between $T_A =$

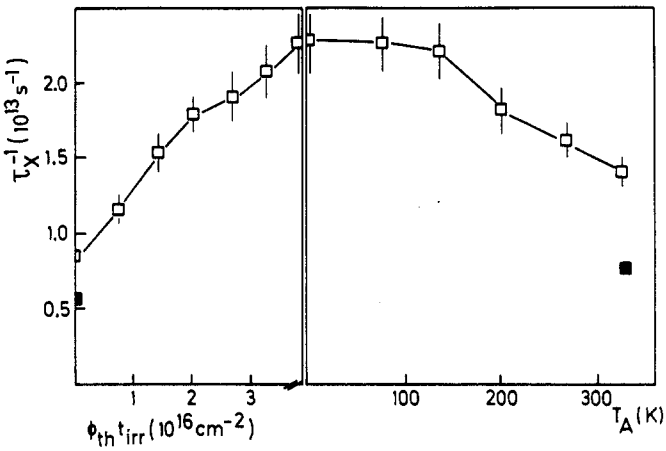


**Figure 6.** Scattering rate $\tau_X^{-1}$ of InAs ($n_0 = 6.8 \times 10^{16}$ cm$^{-3}$) versus thermal neutron dose $\Phi_{th}t_{irr}$ and annealing temperature $T_A$. The $\tau_X^{-1}$-value due to ionised impurity scattering has been calculated for the unirradiated and the transmutation-doped sample using (8) (see full squares). The difference between the measured and the calculated values of the starting material may be attributed to inhomogeneous doping [7].

170 K and $T_A = 270$ K. However, comparing the measured scattering rate $\tau_\rho^{-1}$ ($T_A =$ 350 K) and the calculated value (full square in figure 5) a manifest contribution to $\tau_\rho^{-1}$ of irradiation defect scattering is to be noted even after heating the samples to the highest annealing temperature used in our experiments. It therefore becomes clear that complete annealing cannot be achieved by heating the sample to 350 K. These observations are in good agreement with damage studies by Lindsay and Banbury [14] on InAs containing isolated point defects due to 0.4–2 MeV electron irradiation at 80 K. However, whereas these workers attribute the annealing below 250 K to displaced As atoms, we believe the (n, $\gamma$)-induced In vacancy to be the most common type of defect in our specimens (see below).

Figure 6 shows the scattering rates $\tau_X^{-1}$ as calculated from the Dingle temperature versus thermal neutron dose and annealing temperature. Before irradiation this value exceeds the corresponding transport scattering rate $\tau_\rho^{-1}$ by a factor of approximately 8. The theoretical value for $\tau_X^{-1}$ based on the scattering on singly ionised impurities can be calculated in a similar way to equation (7) [9]:

$$\frac{1}{\tau_{X,ii}} = \left(\sum_i (N_i Z_i^2 e^4) m^* \Big/ 24\pi^3 (\varepsilon_0 \varepsilon_s)^2 \hbar^3 n\right) \frac{1}{2} \frac{b^2}{1+b}. \tag{8}$$

The results of (8) according to the conduction electron densities observed in the measurements before irradiation and after heating the sample to $T_A = 350$ K are inserted in figure 6. The difference between the calculated and the measured scattering rates $\tau_X^{-1}$ of the unirradiated sample may be attributed to inhomogeneities in the doping profile of the starting material, leading to an increase in the Dingle temperature [7, 15]. Similar to the transport scattering rate $\tau_\rho^{-1}$, significant annealing takes place in the temperature range below room temperature but complete annealing is not possible by heating the specimens to 350 K.

Owing to the large differences in the capture cross sections $\sigma$ of In and As it is the In sublattice which is mainly damaged by the irradiation of InAs with thermal neutrons. The (n, $\gamma$)-induced In vacancies and the corresponding interstitials are therefore believed

to be the most common types of defect created in the experiments described above. It is suggested from positron annihilation experiments [16] that in InAs the In vacancy may be neutral or negatively charged. Experimentally only a small difference between the carrier introduction rate during low-temperature thermal neutron irradiation and the calculated transmutation rate has been observed (see figure 3). Furthermore the resistivity of the samples decreased subsequent to the irradiation to its final value, showing a time constant equal to that of the $^{116}$In $\beta$-decay. These findings may most easily be explained assuming that, after being transmuted to $^{116}$Sn, the interstitials act as singly ionised donors, whereas the In vacancies should most probably be neutral.

Low-temperature neutron irradiation of InSb—a material very similar to InAs—leads, in sharp contrast with the results reported above, to a decrease in the conduction electron density in spite of the transmutation doping by donors and a large increase in the resistivity as a function of the thermal neutron dose [5]. From the analysis of these data it was deduced that in InSb the In vacancy (again being the most common irradiation defect) should act as a trapping centre for two electrons. Furthermore the scattering rates $\tau_\rho^{-1}$ and $\tau_X^{-1}$ of InSb were found to increase much more strongly as a function of the neutron dose compared with InAs of the same initial carrier density. This discrepancy may be understood assuming the In vacancy to be doubly charged in InSb and neutral in InAs because in this case in InAs the most common type of defect does not contribute to the otherwise most important scattering mechanism.

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