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Inelastic scattering time of InP after fast neutron irradiation at low temperature

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Abstract. The positive magnetoconductivity $\Delta\sigma(B)$ of InP near the metal–insulator transition (MIT) was measured as a function of the conduction electron concentration and the compensation ratio. Starting deep within the metallic regime the charge carrier density of the sample was reduced stepwise below the critical value of the MIT by fast neutron irradiation due to the acceptor-like nature of the irradiation defects. Up to being exposed to a fast neutron dose of $\Phi t_f = 1.4 \times 10^{16}\,\mathrm{cm}^{-2}$ the sample behaved in a metallic fashion, and subsequent to each irradiation step the inelastic scattering time was deduced from the slope of the $\Delta\sigma = f(B^2)$ curves in the temperature range $2\,\mathrm{K} < T < 6\,\mathrm{K}$. The temperature dependence of the inelastic scattering time was found to vary as T^{-1}, in agreement with a theory developed by Isawa. After the radiation-induced crossover into the insulating regime the $\Delta\sigma \propto B^2$ dependence was still observed. In this case the temperature dependence of the negative magnetoresistance was of the form $\Delta\rho/\rho \propto T^{-\alpha}$ with $\alpha = 1.26$. The influence of the compensation ratio on the measured properties is discussed.

1. Introduction

At low temperatures the conductivity σ_0 of doped semiconductors in the barely metallic regime is dominated by elastic scattering of the conduction electrons by ionized impurities. This process is generally described by the Boltzmann theory, assuming that the momentum of the scattered electron decays exponentially after the scattering event:

$$\sigma_0 = \frac{\tau_0 n e^2}{m^*} \tag{1}$$

where τ_0 is the elastic scattering time, n is the charge carrier density and m^* is the effective mass of the carriers. This picture, however, is known to be incomplete since it neglects the interference of coherently backscattered partial waves. In reality, this process has to be taken into account as long as the coherence is not destroyed by phase-breaking scattering processes, i.e. for times shorter than the dephasing time τ_Φ, which in three-dimensional samples is, to a very good approximation, equal to the inelastic scattering time τ_i [1]. It leads to a decrease of the conductivity below its value according to the Boltzmann theory (see, e.g. [2]). However, the effect of coherent backscattering may be suppressed by a magnetic-field induced phase shift between the electron wavefunctions. The deviation from the usual Boltzmann theory is therefore easily observed by means of the positive magnetoconductivity in weak magnetic fields.

A theoretical picture of the positive magnetoconductivity in three-dimensional systems was developed by Kawabata [3]. Three conditions must be met to apply the theory: (i)

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$(eB\tau_0)/m^* < 1$, (ii) $l_0 < \sqrt{\hbar/(eB)}$, and (iii) $k_F l_0 > 1$ where k_F and l_0 are the Fermi wavevector and the elastic mean free path respectively. The last inequality, known as the Ioffe–Regel condition, restricts the validity of the theory to metallic materials. In general, most of the experimental work was performed using degenerate semiconductors.

In SI units the magnetic-field induced increase of conductivity $\Delta\sigma$ is given by

$$\Delta\sigma = \frac{e^2}{2\pi^2\hbar}\sqrt{e/\hbar}\,F(\delta)\sqrt{B} \tag{2}$$

with $\delta = \hbar/4eBD\tau_i$ and D the diffusion constant. Strictly, $F(\delta)$ is the limit of an infinite series. However, for weak magnetic fields, i.e. for $\delta > 1$, (2) can be simplified. Using $F(\delta) \cong \frac{1}{48}\delta^{-3/2}$ the positive magnetoconductivity may be written as

$$\Delta\sigma = \frac{\sigma_0}{12\sqrt{3}}\left(\frac{\tau_i}{\tau_0}\right)^{3/2}\left(\frac{e\tau_0}{m^*}\right)^2 B^2. \tag{3}$$

It is obvious from this formula that the measurement of the positive magnetoconductivity in low fields provides a tool for the determination of the inelastic scattering time. In metallic InP experiments of this type were performed by Finlayson and Mehaffey [4] and more recently by Biskupski and Bouattou [5].

Only a few investigations deal with the low-temperature magnetotransport properties of disordered insulating material, the conductivity of which is determined by the variable range hopping mechanism. A theory by Sivan and co-workers [6], however, predicts a positive magnetoconductivity for strongly disordered insulators obeying a B^2 law. Experimentally this theory is supported by the findings of Tremblay and co-workers [7] in GaAs and by Finlayson and Mehaffey in InP [4].

The temperature dependence of the inelastic scattering rate in three-dimensional disordered metals was analysed by Isawa [8], resulting in a linear dependence at low temperature. Above a crossover temperature determined by the elastic scattering time ($T_c = C/\tau_0$; C = constant = 1.267×10^{-12} K s) the linear regime is superposed by a $T^{3/2}$ dependence.

The aim of this work was to provide data on the temperature dependence of the inelastic scattering time of InP as a function of the conduction electron density and the degree of compensation. Starting with a degenerate InP crystal the charge carrier concentration of the sample was stepwise reduced by the introduction of irradiation defects, which in InP are known to be acceptor-like, e.g. [9]. In this way the positive magnetoconductivity of the same InP specimen was measured for different values of conduction electron density and compensation ratio on both sides of the metal insulator transition.

2. Experimental procedure

The starting material for the experiment was a nominally undoped InP wafer exhibiting a conduction electron density of $n_0 \approx 6 \times 10^{15}\,\mathrm{cm}^{-3}$ at room temperature. The samples were cut into rectangular parallelepipeds of $5 \times 2.5 \times 0.5\,\mathrm{mm}^3$.

Since the Kawabata theory of weak localization is applicable for degenerate material only, the specimens were n-doped by means of the neutron transmutation doping (NTD) technique to a donor concentration of $N_D = 2 \times 10^{17}\,\mathrm{cm}^{-3}$, i.e. a level lying well within the metallic regime. The NTD was performed using the RP-R2 irradiation position of the Munich

Research Reactor, characterized by a thermal neutron flux of $\Phi_{th} = 9 \times 10^{12}\,cm^{-2}\,s^{-1}$ and a ratio of thermal-to-fast ($E > 0.1\,MeV$) neutron fluxes of $\Phi_{th}/\Phi_f \approx 15$. In all In-containing compound semiconductors the most important transmutation reaction is $^{115}In(n,\gamma)^{116}In \xrightarrow{\beta^-} {}^{116}Sn$ producing tin donors in the In sublattice. It is well known, however, that the displacement energies of InP are among the lowest for all materials, i.e. $E_d < 10\,eV$ for In and for P atoms as well [10]. Hence the neutron capture during the NTD reaction is connected with the production of a close Frenkel-pair due to the recoil energy of typically 50 eV [11] transferred to the ^{116}In during the γ emission. Subsequent to the NTD, and a waiting time of about four weeks necessary for the decay of the induced radioactivity, the specimens were thermally annealed in order to remove the irradiation defects. The annealing was carried out in an atmosphere of inert gas at a temperature of 870 K. During annealing the specimens were covered by two other InP wafers forming a sandwich-like structure. In this way a possible loss of P due to its low vapour pressure [12] was avoided. Finally, Ohmic contacts were attached to the sample surface by soldering with pure In.

Before irradiation and subsequent to each dose level the conduction electron density and the compensation ratio were determined by means of resistivity and Hall-effect measurements at 77 K [13]. These data were taken using the van der Pauw method [14] in order to avoid systematic errors due to the sample geometry.

The irradiations were performed at the low-temperature irradiation facility of the Munich Research Reactor [15]. This facility allows in-core reactor irradiations at $T < 6\,K$ using an empty fuel element position and subsequent transfer of irradiated specimens into a measuring cryostat without intermediate warming up. The thermal and fast neutron fluxes are $\Phi_{th} \approx \Phi_f \approx 3 \times 10^{13}\,cm^{-2}\,s^{-1}$. The maximum fluence in the experiments presented here was $\Phi t_f = \Phi t_{th} \approx 2.1 \times 10^{16}\,cm^{-2}$. From these data the additional transmutation doping level introduced during low-temperature irradiation is calculated to be $\Delta N_{NTD} = 8.4 \times 10^{16}\,cm^{-3}$.

The measuring cryostat was equipped with a superconducting coil and a controllable furnace needed for the measurement of the compensation ratio at 77 K. By pumping the He bath and heating the sample holder the measuring temperature was varied between 2–6 K. For the temperature measurement Allen Bradley carbon resistors were installed in the sample holder in a position equivalent to the one of the specimens themselves. These resistors are known to be sensitive to reactor irradiation [16] only after being exposed to very high neutron doses. In the experiments described here their temperature characteristic was therefore regarded to be unaffected by the irradiation.

3. Results and discussion

Figure 1 shows the electrical conductivity at $T = 80\,K$ as a function of the fast neutron dose. In figure 2 the corresponding curve is shown for the conduction electron density n. Within the error bars the carrier concentration decreases linearly with a rate of $\Delta n/\Delta(\Phi t_f) = (-6.46 \pm 0.63)\,cm^{-1}$. It is noteworthy that this decrease in carrier density is found in spite of the unavoidable transmutation doping of the samples during low-temperature irradiation by the fraction of thermal neutrons in the reactor spectrum. Similar effects have already been observed during low-temperature transmutation doping of InSb [17]. They are to be attributed to the overcompensation of the introduced donors by charged acceptor-like lattice defects.

The proper characterization of the sample material requires a knowledge of the compensation ratio k, to be determined using the data shown in figures 1 and 2:

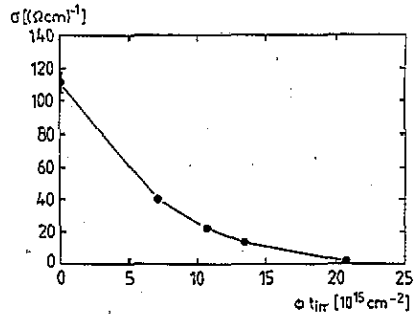

Figure 1. Electrical conductivity $\sigma(T = 80\,\mathrm{K})$ of InP as a function of the fast neutron dose Φt_f. The conduction electron density of the non-irradiated material was $n_0(T = 80\,\mathrm{K}) = 1.64 \times 10^{17}\,\mathrm{cm}^{-3}$.

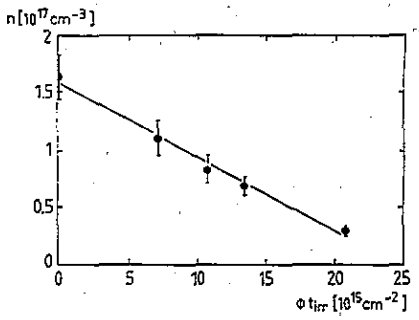

Figure 2. Conduction electron concentration $n(T = 80\,\mathrm{K})$ of InP as a function of the fast neutron dose Φt_f.

$$k = \frac{N_A}{N_D} = \frac{(N_D + N_A) - n}{(N_D + N_A) + n} \tag{4}$$

where N_A and N_D are the acceptor and donor density. The total ion density $N_D + N_A$ was determined from the comparison of the measured elastic scattering rate with the theoretical ionized impurity scattering rate, e.g. [18]

$$\frac{ne^2}{\sigma m^*} = \frac{\sum_i (N_i Z_i^2) e^4 m^*}{24\pi^3 (\epsilon_0 \epsilon_s)^2 \hbar^3 n} \left(\ln(1+b) - \frac{b}{1+b} \right). \tag{5}$$

In (5) the parameter $b = (2k_F R_{TF})^2$ is used; k_F is the Fermi wavevector, R_{TF} the Thomas Fermi screening length, Ze the charge of the ion and $\epsilon_s = 12.56$ [19] the static dielectric constant of InP. For an unambiguous interpretation of (5), however, the charge of the various types of defects has to be known. Because of the careful thermal treatment it seemed reasonable to assume the defect structure of the starting material for the low-temperature irradiation to be composed from singly charged defects only, i.e. from ionized tin atoms due to the NTD and shallow acceptor impurities. After fast neutron irradiation, on the other hand, multiple charged defects and amphoteric clusters are known to be present in the sample [20]. Therefore in this case the compensation ratio has to be interpreted as the fraction of trapped electrons as compared to the donor density. In figure 3 the compensation ratio is shown as a function of the fast neutron dose and the conduction electron density at $T = 80\,\mathrm{K}$ respectively. It starts with a rather low value of 18% and is increased up to 89% after exposure of the samples to the maximum dose of $\Phi t_f = 2.1 \times 10^{16}\,\mathrm{cm}^{-2}$.

In the InP specimens the positive magnetoconductivity was observed in unirradiated material and after each irradiation step. According to the theory $\Delta\sigma/\sigma_0$ was found to vary with B^2 for weak magnetic fields. From a typical result, as shown in figure 4, it can be seen that the effect is more pronounced for lower temperatures.

For the discussion of the data it has to be noted that in irradiated samples the Ioffe–Regel condition $k_F l_0 > 1$ is fulfilled only up to a fast neutron dose of $\Phi t_f \approx 1.1 \times 10^{16}\,\mathrm{cm}^{-2}$, i.e. approximately half the maximum dose (see table 1). After being exposed to $\Phi t_f = 1.4 \times 10^{16}\,\mathrm{cm}^{-2}$ the specimen is characterized by $k_F l_0 = 0.55$. Nevertheless, it behaves in a nearly metallic fashion, as can be seen from the temperature dependence of the zero-field conductivity (figure 5). This contradiction to the Ioffe–Regel condition is most

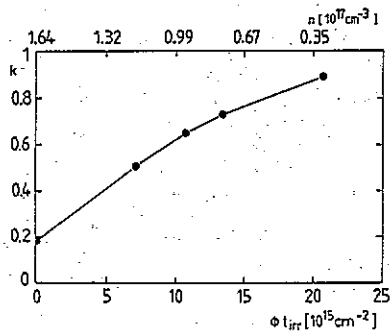

Figure 3. Compensation ratio k as a function of the fast neutron dose Φt_f and the conduction electron density $n(T = 80\,\mathrm{K})$.

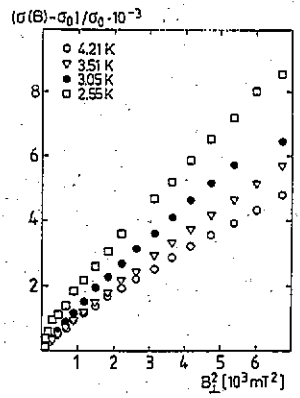

Figure 4. Normalized magnetoconductivity $\Delta\sigma/\sigma_0$ plotted against the square of the magnetic field $B_\perp^2$ for different temperatures. The data were taken after fast neutron irradiation up to $\Phi t_f = 1.4 \times 10^{16}\,\mathrm{cm}^{-2}$.

probably caused by the drastic decrease of the elastic mean-free path l_0 due to the irradiation defects. A further argument for the metallic nature of the sample, despite that $k_F l_0 < 1$, is the fact that the charge carrier density falls below the critical value according to the Mott criterion—$n_c \approx 3.4 \times 10^{16}\,\mathrm{cm}^{-3}$—only after application of the maximum neutron dose leading to a clearly insulating characteristic of the specimen (see figure 5). However, the B^2 behaviour of the positive magnetoconductivity was observed in any case and particularly in the insulating regime.

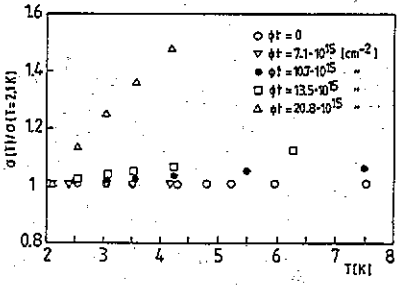

Figure 5. Temperature dependence of the normalized zero-field conductivity $\sigma(T)/\sigma(T = 2.1\,\mathrm{K})$ for InP after exposure to different fast neutron doses. The corresponding values for the conduction electron density and the compensation ratio may be extracted from figure 3.

After the determination of the elastic scattering time τ_0 by means of (1) the inelastic scattering time τ_i was calculated from the slopes of the magnetoconductivity curves according to (3). Since after exposure to the maximum irradiation dose the specimen was clearly on the insulating side of the MIT this evaluation was justified for irradiation doses of $\Phi t_f \leqslant 1.4 \times 10^{16}\,\mathrm{cm}^{-2}$ only. In figures 6 and 7 the elastic and the inelastic scattering times at $T = 4.2\,\mathrm{K}$ are shown as a function of the fast neutron dose and the conduction electron density $n(T = 4.2\,\mathrm{K})$ respectively. As was to be expected the inelastic scattering time in the unirradiated InP turns out to be approximately two orders of magnitude larger than the elastic one; but whereas the elastic scattering rate is strongly decreased due to the introduction of irradiation defects, τ_i is only little affected by neutron irradiation. This fact can qualitatively be understood in the Kawabata theory: (3) may be written as

$$\tau_i = \text{constant} \times \left(\frac{\Delta\sigma}{\sigma_0}\right)^{2/3} \tau_0^{-1/3}. \tag{6}$$

It is known from the theory [21] that the coherent backscattering intensity, being the reason for the positive magnetoconductivity, is independent of the number of intermediate elastic scattering events. In fact, for the same temperature $\Delta\sigma/\sigma_0$ was found to vary only little with an increasing neutron dose, i.e. to be almost independent of the elastic scattering time. Therefore a rough estimation results in $\tau_i \propto \tau_0^{-1/3}$. It is plausible from this consideration that the inelastic scattering time increases slightly, in spite of the reduced charge carrier density in irradiated material. This finding is in contradiction to the results of [4], probably gained from material with a constant degree of compensation.

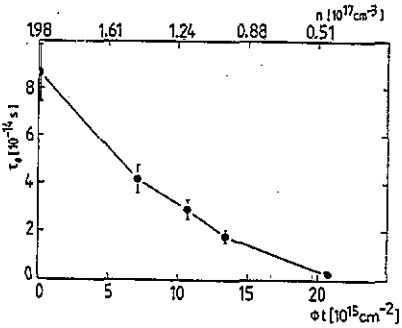

Figure 6. Decrease of the elastic scattering time $\tau_0(T = 4.2\,\text{K})$ as a function of the fast neutron dose Φt_f and the conduction electron density $n(T = 4.2\,\text{K})$.

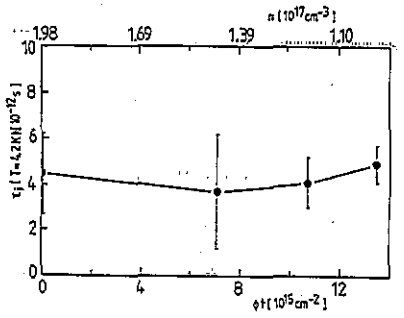

Figure 7. Inelastic scattering time τ_i as a function of the fast neutron dose Φt_f and the conduction electron density $n(T = 4.2\,\text{K})$. The τ_i data were evaluated from the slopes of the magnetoconductivity curves according to the Kawabata theory.

Figure 8 shows in a double logarithmic plot the temperature dependence of the inelastic scattering time using the fast neutron dose, or in other words the charge carrier density (see figure 2), as a parameter. A function of the form $\tau_i^{-1} = AT^\kappa$ (A = constant) was fitted to each set of data using a least-squares routine. The results obtained in the best fits are given in table 1 together with the most important material parameters. As mentioned above the Isawa theory [8] predicts a value of $\kappa = 1$ at low temperatures. It is shown in table 1 that for the non-irradiated starting material the fit resulted in $\kappa = 1.28$. The most probable reason for this higher value is that the crossover temperature T_C separating the $\tau_i^{-1} \propto T$ from the $\tau_i^{-1} \propto T^{3/2}$ regime is only $T_C = 14.3\,\text{K}$ due to the comparatively large elastic scattering time of the non-irradiated sample. After exposure to the first two irradiation doses, however, the Ioffe–Regel condition is still fulfilled ($k_F l_0 > 1$) and in agreement with the theory κ is found to be close to 1. The same behaviour is observed even after subsequent irradiation ($\Phi t = 1.4 \times 10^{16}\,\text{cm}^{-2}$). Although in this case the Ioffe–Regel condition is violated ($k_F l_0 = 0.55$) it can be seen from figure 5 that the specimen does not behave like a typical insulator.

As mentioned above a negative quadratic low-field magnetoresistance was observed earlier in strongly disordered insulating GaAs [7]. According to this work the temperature dependence of the magnetoresistance may be described by

$$\frac{\Delta\rho}{\rho} = \frac{\rho(B) - \rho(0)}{\rho(0)} = yT^{-\alpha}B^2 \tag{7}$$

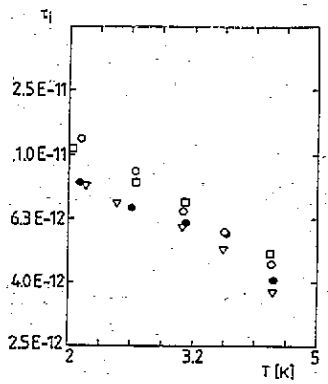

Figure 8. Double logarithmic plot of the temperature dependence of the inelastic scattering time τ_i of InP after exposure to different fast neutron doses. (open circles: $(\Phi t)_f = 0$; triangles: $(\Phi t)_f = 7.1 \times 10^{15}\,\mathrm{cm}^{-2}$; full circles: $(\Phi t)_f = 10.7 \times 10^{15}\,\mathrm{cm}^{-2}$; squares: $(\Phi t)_f = 13.5 \times 10^{15}\,\mathrm{cm}^{-2}$).

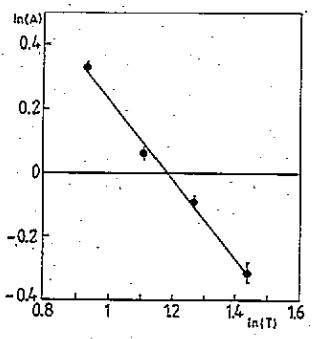

Figure 9. Double logarithmic plot of $A = \Delta\rho/(\rho B^2)$ against temperature T. The data were obtained after exposure to the maximum dose of $\Phi t_f = 2.1 \times 10^{16}\,\mathrm{cm}^{-2}$. The sample characteristics are $n(T = 80\,\mathrm{K}) = 3.0 \times 10^{16}\,\mathrm{cm}^{-2}$ and $k = 0.89$.

Table 1. Important parameters of the irradiated InP samples.

Φt_f (cm^{-2})	n (cm^{-3})	$k_F l_0$	k	κ
0	1.98×10^{17}	4.37	0.18	1.28
7.1×10^{15}	1.39×10^{17}	1.68	0.51	1.07
1.1×10^{16}	1.14×10^{17}	1.03	0.65	0.92
1.4×10^{16}	0.92×10^{17}	0.55	0.73	0.98
2.1×10^{16}	0.47×10^{17}	0.06	0.89	—

with y and α being constants. In [7] a value of $\alpha = 1.22$ was evaluated from temperature-dependent magnetoresistance measurements. Figure 9 shows a double logarithmic plot of $A = \Delta\rho/(\rho B^2)$ against T. In good agreement with [7], a least-squares fit results in a value of $\alpha = 1.26$.

In general we may conclude that the results presented above agree with the theoretical predictions [3, 6, 8] and with experiment [4, 5, 7] as well. The neutron irradiation made it possible to observe the positive magnetoconductivity in the same sample on both sides of the metal insulator transition as a function of conduction electron density and compensation ratio. Since the role of the latter parameter is not yet quite clear a subsequent experiment using a sample with fixed carrier density and variable compensation ratio will be performed. In a further experiment it is intended to study in more detail the transition from weak to strong localization by magnetotransport and temperature-dependent conductivity measurements in the insulating regime.

Acknowledgments

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