Superheated free-electron gas in n-GaAs: the role of X-valley electrons in optical nonlinearities at 10.6 µm.

G. Shkerdin[1], J. Stiens[2], R. Vounckx[2]

[1] Institute of Radio Engineering and Electronics of RAS, Vvedensky Square 1, R-141120 Fryazino (Moscow region), Russia.

[2] Vrije Universiteit Brussel, Lab for Micro- and Optoelectronics, Electronics Department, Pleinlaan 2, B-1050 Brussels, Belgium.

By optically heating free electrons in highly doped n-GaAs up to electron temperatures of about 1000K with short and very powerful mid- or far infrared light pulses, electrons can be redistributed between three different valleys: degenerated nonparabolic Γ-, anisotropic L- and X-valley. As the effective masses and the mobilities of the free electrons in the three valleys are very different, the population modulation induces changes in the dielectric permittivity function and hence optical nonlinearities[1,2]. We derived a novel appropriate quantum-mechanical model, which describes the impact of the different valley geometries and the many different intervalley deformation potentials.

1. Introduction

In n-GaAs, the separation between the Γ- and X-valleys is considerably large (almost 500 meV) and usually the influence of the X-valley electrons on GaAs optical properties is neglected. Powerful mid-infrared (MIR) light pulses, however, can increase the electron temperature of highly doped n-GaAs up to about 1000K. In this superheated situation a considerable amount of conduction electrons is already transferred to the X-valley. Therefore existing opto-electronic models have to be corrected at high electron temperatures when free electron gas is distributed between three different valleys: central Γ-valley, the L-valley and the X-valley.

2. The model of GaAs optical properties calculation

To treat the effect of free electron absorption in GaAs, we use a rigid quantum mechanical approach. In this paper we take into account the distribution of conduction electrons between the nonparabolic Γ-valley, the ellipsoidal L- and X-valleys and absorption mechanisms connected with electron transitions between all equivalent and nonequivalent energy valley minima.

By using Drude's model we can write down the expression for the dielectric permittivity ε_e at medium infrared wavelength:

$$\varepsilon_e(\omega)=\varepsilon_1\left(1-\frac{\omega_{p,\Gamma}^2+\omega_{p,L}^2+\omega_{p,X}^2}{\omega^2}+i\left(\frac{\omega_{p,\Gamma}^2}{\omega^3\tau_\Gamma}+\frac{\omega_{p,L}^2}{\omega^3\tau_L}+\frac{\omega_{p,X}^2}{\omega^3\tau_X}\right)\right) \quad (1)$$

where $\omega_{p,\Gamma,L,X}$, $n_{\Gamma,L,X}$ and $m_{\Gamma,L,X}$ are the plasma frequency, the electron concentration and optical effective mass of the $\Gamma-$, L- and X- valley electrons, respectively and ω is the electromagnetic wave angular frequency. The values $\tau_{\Gamma,L,X}$ are directly connected with the absorption coefficients by $\Gamma-$, L - and X-valley electrons and can be determined from the solution of the photon kinetic equation described in Ref.[1,2]. The total absorption coefficient α_e by Γ, L and X-electrons is given by the expression:

$$\alpha_e = 2\frac{\omega}{c}\operatorname{Im}\sqrt{\varepsilon_e(\omega)} \tag{2}$$

The absorption mechanisms in the X-valley are subdivided between intravalley and intervalley ones. The main intravalley mechanisms are due to the electron interaction with impurities and optical phonons. The intervalley absorption mechanisms by free electrons are induced by electron interaction with $\Gamma-L$, $L-L$, $X-\Gamma$, $X-L$ and $X-X$ intervalley phonons. These mechanisms are described by the five intervalley deformation potentials: $\Lambda_{\Gamma L}$, Λ_{LL}, $\Lambda_{\Gamma X}$, Λ_{LX} and Λ_{XX}.

In order to calculate the dependence of the absorption coefficient α_e or the dielectric permittivity $\varepsilon_e(\omega)$ on the laser intensity W we have to know the dependence of electron temperature t_e on W. This dependence can be found from the solution of the stationary balance equation:

$$\alpha_e W = P \tag{3}$$

Here P is the total power transferred to the lattice by the heated free electron gas during the energy relaxation process. The stationary regime corresponds to a situation where the optical pulses are much longer than the relaxation times of the involved nonlinear mechanisms and short enough to avoid any thermal heating of the lattice. The estimations show that optical pulses with nanosecond duration satisfy the stationary regime conditions for $t \le 1000K$.

The total power transferred to the lattice is given by:

$$P = P_\Gamma + P_L + P_X + P_{\Gamma L} + P_{L\Gamma} + P_{\Gamma X} + P_{X\Gamma} + P_{LX} + P_{XL} \tag{4}$$

where $P_{\Gamma,L,X}$ is the power transferred to the lattice by Γ-, L- and X- valley electrons, respectively, taking into account only intravalley and equivalent intervalley transitions. All other mixed terms describe the power transferred to the lattice as a result of nonequivalent intervalley $\Gamma-L$, $\Gamma-X$ and $L-X$ phonons emission by conduction electrons. Substitution of Exp.(4) into Eq.(3) leads to the equation for the electron temperature dependence $t_e(W)$ and using Exp.(1), to the final intensity dependent absorption coefficient and dielectric permittivity.

3. Results of numerical calculations and discussion

The material parameters of GaAs used in the numerical calculations are given in Ref.[1-3]. For the intervalley deformation potentials, the values copied from literature scatter as much as about one order of magnitude [3,4]. This leads to a large uncertainty of intervalley transition contributions to free electron absorption process.

The influence of X-valley electrons on the absorption coefficient and dielectric permittivity is proportional to X-electron concentration n_X for small values of n_X. One can calculate that the relative concentration of X-electrons at $t = 1000K$ is about 9% for $n_0 = 7.6\times10^{18} cm^{-3}$. The optical effective mass of X-electrons is considerably larger than that of L-electrons ($m_X / m_L \approx 2.87$) and Γ-electrons ($m_X / m_\Gamma \approx 3.88$ for $t_e = 1000.K$ and $n_0 = 7.6\times10^{18} cm^{-3}$). Therefore, the transfer of electrons from the Γ-

valley to subsidiary valleys leads to a drastic decrease of the effective plasma frequency $\omega_{p,ef} = \sqrt{\omega_{p,\Gamma}^2 + \omega_{p,L}^2 + \omega_{p,X}^2}$. The larger optical effective mass of X-electrons results in a stronger influence of these electrons on the $\omega_{p,ef}$ value and the absorption coefficient. In this sense the influence of a small X-electron concentration is amplified by the factor m_X / m_L compared to the influence of the same small concentration of L- electrons.

The main intervalley contribution to the absorption processes connected with the X- valley is given by $L-X$ intervalley transitions. This is explained by the large density of states in the L-valley for electron energies close to the X- valley bottom and the large difference of electron optical effective masses in X-and L-valleys. The influence of intervalley deformation potential constants on the absorption coefficient is especially large at high electron temperatures. The uncertainty interval for the $\Lambda_{\Gamma X}$, Λ_{LX} and Λ_{XX} coefficients leads to an absorption uncertainty of (16-38)% at electron temperatures $(700K \le t_e \le 1000K)$. Hence, the absorption contribution of X-valley electrons is considerable and has to be definitely taken into account at $t_e \sim (900-1000)K$.

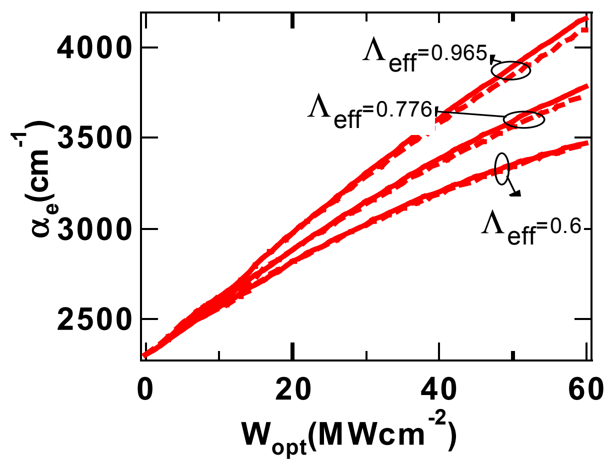

Fig. 1 The intensity dependent absorption coefficient for different effective deformation potential values expressed in 10^9eVcm^{-1}. Doping concentration n_o=7.6x10^{18}cm^{-3}.

In experiments, only the intensity dependent absorption coefficient can be measured. Generally $\alpha_e(W)$ depends on all X-valley intervalley deformational potentials $\Lambda_{\Gamma X}$, Λ_{LX} and Λ_{XX} separately. Increase or decrease of these values leads to the appropriate increase or decrease of the absorption coefficient. As the main contribution to absorption coefficient is given by Λ_{LX} value we can suggest that the influence of X valley on $\alpha_e(W)$ dependence can be approximated by a following combination: $\tilde{\Lambda} = \sqrt{\Lambda_{LX}^2 + a(\Lambda_{\Gamma X}^2 + \Lambda_{XX}^2)}$ where a is a small value and depends on the doping concentration. This suggestion is confirmed by numerical calculations. Dependencies of $\alpha_e(W)$ are plotted in Fig.(1) for the doping concentration $n_0 = 7.6\times10^{18} cm^{-3}$ and different values of $\Lambda_{\Gamma X}$, Λ_{LX} and Λ_{XX} that are chosen to satisfy the condition: $\tilde{\Lambda} \approx (0.965, 0.776, 0.6)\times10^9 eV/cm$ for the upper, medium and lower curves, accordingly (here $a = 0.1$). Analogous results for $n_0 = 2.5\times10^{18}\,cm^{-3}$ are obtained with $a = 0.04$ and for $n_0 = 1\times10^{18}\,cm^{-3}$ with $a = 0.025$.

Therefore, experimental measurements of $\alpha_e(W)$ can provide only the value of $\tilde{\Lambda}$ if deformation potential constants $\Lambda_{\Gamma L}$ and Λ_{LL} are known. For this reason Λ_{LX} evaluations become more accurate for its larger value, smaller doping concentration and for the cases when $\Lambda_{\Gamma X}$ and Λ_{XX} are known more precisely.

The analysis of the real part of dielectric permittivity versus intensity shows that for a doping concentration $n_0 \sim 7\times10^{18}\,cm^{-3}$ the influence of X-valley electrons is very small for relatively small MIR intensity $W \le (6-8)MW/cm^2$. The inclination angle of $\mathrm{Re}(\varepsilon(W))$ dependence and the nonlinear index of refraction n_2 at these intensities depends on the Λ_{LL} value (see also Ref.[2]). For larger intensities $W \sim 40MW/cm^2$ when electron temperature $t_e \sim 900K$ the influence of X-valley electrons becomes considerable and has to be taken into account.

4. Conclusions

The analytic quantum mechanical model describing absorption of medium infrared light by superheated free electrons distributed between degenerate nonparabolic Γ–valley and anisotropic L- and X- valleys in highly doped n-GaAs was developed. All particle interactions giving the main contribution to absorption coefficient have been taken into account (the intravalley electron-optical phonon and electron-impurity interactions and the interactions of electrons with different types of intervalley phonons). The proper model describing the power transferred to the lattice from the heated electron gas was also elaborated. These models enable to calculate the intensity dependent MIR absorption coefficient and dielectric permittivity. It was shown that X-valley electrons give a considerable contribution to nonlinear dielectric permittivity and MIR absorption coefficient α_e for electron temperatures $t_e \ge (900-1000)K$. The influence of X valley on MIR absorption coefficient for a fixed doping concentration can be approximately described by an effective deformation potential $\tilde{\Lambda} = \sqrt{\Lambda_{LX}^2 + a(\Lambda_{\Gamma X}^2 + \Lambda_{XX}^2)}$ where a depends on the doping concentration.

Acknowledgments

This work was partially funded by the VUB-GOA (Concerted Research Actions), OZR-VUB and FWO-V.

References

1. G. Shkerdin, J. Stiens, R. Vounckx, Comparative study of the intra- and intervalley contributions to the free-carrier induced optical nonlinearity in n-GaAs, J. Appl. Phys., vol. 87, pp.3807-3818, 1999.
2. G. Shkerdin, J. Stiens, R. Vounckx, A Multi-Valley Model for Hot Free-Electron Nonlinearities at 10.6 μm in Highly Doped n-GaAs, EPJ- Appl. Phys. vol. 12, pp.169-180, 2000.
3. S.Adachi, GaAs, AlAs, and $Al_xGa_{1-x}As$: Material parameters for use in research and device applications, Appl.Phys., vol. **58**, pp.R1-R29, 1985.
4. S.Zollner, S.Gopalan, M.Cardona, Microscopic theory of intervalley scattering in GaAs: k dependence of deformation potentials and scattering rates, J. Appl. Phys., vol. 68, pp.1682-1693, 1990.