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PHOTOCONDUCTIVITY IN GaAlAs:Si PROVES NEGATIVE U OF DX CENTERS

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The analysis of the temperature dependence of the photoconductivity, amplitude in doped GaAlAs, provides a simple and convincing proof of the negative sign of the Hubbard correlation energy U, strictly speaking of the two-electron nature of the thermal emission process from DX centers. The proof is based on a comparison of the emission activation energy measured per emission event (DLTS) with that measured per electron.

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GaAlAs doped with donors exhibits extremely long relaxation times of the optically excited electrons and a strong photoconductivity effect is observed [1, 2]. It is possible to change and to control the position of the quasi-Fermi level within a wide range of energies. There is no doubt that besides a shallow donor state there is a deep state (DX) of the same donor and that these states are separated by an effective barrier [2]. There is, however, a controversy on the nature of the DX state and on the character of the barrier. The theoretical models calculated by Chadi and Chang [3] and by Dabrowski et al. [4] predict the existence of a two-electron deep state (negative effective Hubbard energy U of the DX state) while the calculations of a similar type made by Yamaguchi et al. show [5] that a neutral D^0 state is the ground state of the relaxed system. There is also no conclusive experimental evidence about the number of electrons captured on the DX state. Though there are many experimental data available [6], most of them can be as well described within the positive U as in the negative U model. Up to now, the only convincing experimental indication validating the negative U model comes from the analysis of the capture and emission kinetics [7–10].

The idea of this paper is based on the comparison of the barrier heights seen from both sides of the barrier. The height seen from the DX state corresponds to the activation energy of the emission process, while that seen from the opposite side corresponds to the capture activation energy. Both quantities can be investigated experimentally. The height of the emission barrier is well determined from DLTS measurements and from investigations of the kinetics of conductivity, and is equal to $E_e = 0.4$ eV [6–8]. It is, however, unclear whether the activation energy of this process corresponds to the emission of a single electron or of two electrons, i.e. whether one needs 0.4 eV per electron or 0.2 eV per electron for thermal ionization.

The character of the capture barrier is much more complicated since it depends on the position of the quasi-Fermi level, which varies during the capture process leading to strongly nonexponential transients. Moreover, the capture process in GaAlAs is additionally complicated by alloy splitting of DX levels [9]. In consequence, a quantitative description of the capture transients is extremely difficult. In this paper, however, we will show that in order to solve the problem of the sign of the Hubbard energy U of the DX centers one need not analyze the capture kinetics in detail. As long as we are interested only in the number of electrons captured on the DX center it is enough to investigate the upper limit of the quasi-Fermi level under illumination.

It is clear that in the case of positive U, when a single electron is captured on the DX center, the quasi-Fermi level extrapolated to ze temperature and/or strong illumination should coincide with the top of the barrier. The energy of the barrier top is higher than the energy of the DX center E_{DX} by the emission activation energy E_e. If one notices that at thermal equilibrium, before illumination, the Fermi energy is equal to the one-electron DX energy E_{DX} (in strongly doped GaAlAs samples), one can conclude that the range of changes of the quasi-Fermi energy under illumination are limited by the height of the emission barrier (0.4 eV).

In the other case, when two electrons are captured on the DX center, the change of the quasi-Fermi energy corresponds to the change of the energy of each of the two electrons which take part in the capture process. Thus, the energy barrier for the capture of two electrons is reduced twice as much as the quasi-Fermi energy is increased. So, in order to get the upper limit of PPC it is enough to increase the quasi-Fermi level by a half of the emission energy, i.e. by 0.2 eV. This is only the half of the energy value required in the first case.

Concluding, the idea of the experiment is based on the assumption that the quasi-Fermi energy extrapolated to low temperature and/or to high illumination corresponds to the energy of the top of the barrier per electron. After comparison with one-electron DX energy E_{DX} it determines the emission activation energy per electron. Finally, the ratio of the emission activation energy (measured by the DLTS for example), i.e. the energy required for one emission event, to the energy barrier for electron emission determined per electron (measured by the upper limit of the quasi-Fermi level) gives the number of electrons which take part in a single emission event.

The experimental results are shown in Fig. 1. The crosses show the posi-

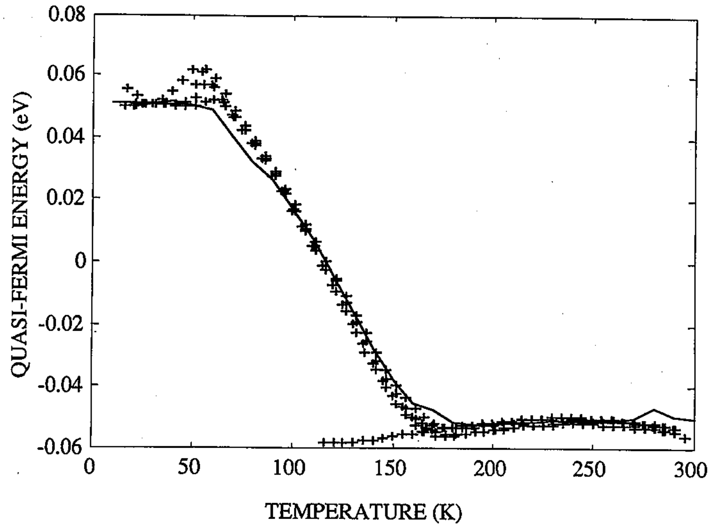

Fig. 1. Quasi-Fermi level as obtained from the conduction electron concentration of an $Ga_{1-x}Al_xAs$:Si sample ($x = 0.31$, $N_D = 2 \times 10^{18}$ cm^{-3}) under illumination by an infrared light emitting diode (dark sample and three different illumination intensity) as a function of temperature — crosses. The theory [7, 9] — solid line.

tion of the quasi-Fermi energy. At high temperature the Fermi level is pinned to the DX energy at about 0.05 eV below the bottom of the conduction band. At 170 K the PPC effect appears and the quasi-Fermi energy increases with decreasing temperature. The increase of the quasi-Fermi energy is practically linear with temperature and the extrapolation to zero temperature gives the upper limit of the quasi-Fermi energy, i.e. the position of the top of the barrier, at 0.150 eV. Thus the resulting value of the emission barrier per one electron is 0.2 eV. But this is just the half of the emission activation energy measured per one event. So, it proves that in the thermal emission process from the DX state two electrons are emitted simultaneously, and consequently, the Hubbard correlation energy is negative, $U < 0$.

The solid line in the figure has been calculated within the model where the alloy splitting of the DX level and of the top of the barrier has been taken into consideration. The details of model are presented elsewhere [7, 9].

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