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Spatial correlations of remote impurity charges: Mechanism responsible for the high mobility of a two-dimensional electron gas

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We have examined the electron mobility μ as a function of the concentration of a two-dimensional electron gas n, in GaAs/$Al_xGa_{1-x}As$ heterostructures. Depending on the method used to vary n, different values of μ can be obtained for the same n and in the same heterostructure. We suggest the origin of this effect lies in the interdonor interactions leading to spatial correlations of remote impurity charges. It results in a significant mobility enhancement at low temperatures. This finding demonstrates the necessity of taking into account many-body effects within the impurity system in different phenomena occurring in highly doped semiconductors.

The problem of Coulomb interactions between charges in various systems has recently attracted a considerable amount of interest. Examples where this phenomenon manifests itself are ordering in an electron gas[1] and in quantum-liquid states associated with the fractional quantum Hall effect.[2] The degree of ordering due to the interactions depends on many external and internal parameters of a system such as, e.g., temperature, magnetic field, strength of interaction, homogeneity of the system. The problem of a coexistence of inherent disorder with the charge-position correlations due to Coulomb interactions has been studied previously in the case of a two-dimensional electron gas (2DEG),[3] charge-density waves on a random alloy, and flux lines in high-temperature superconductors.[4]

In this paper we will concentrate on a different aspect of strong Coulomb correlations represented by impurities in semiconductors. In this situation the positions of charges are restricted to a randomly distributed and fixed set of sites in the matrix lattice. The importance of the interimpurity interactions has been recognized previously in the context of hopping conductivity and the formation of the Coulomb gap in the impurity density of states.[5] Under the condition of a partial occupancy of impurities by electrons, Coulomb interactions among the impurity charges may induce a spatial correlation of positions of these charges.[6] It results in a lowering of the total energy of the system. Moreover, spatially correlated charged impurities are less effective scatterers of the conduction electrons compared to a random distribution of scattering charges. As a consequence, the electron mobility is enhanced making this subject also interesting from a device-oriented point of view.

The purpose of this paper was to answer a question about the existence of spatial correlations of impurity charges in a 2D semiconductor system. To study this effect we used an analysis of the 2DEG mobility μ in modulation-doped heterostructures of GaAs/$Al_xGa_{1-x}As$.[7] The commonly accepted approach to 2DEG scattering in this important semiconductor system neglects many-body effects within the impurity system.

Donors in GaAs and $Al_xGa_{1-x}As$ represent so-called DX centers.[8] Transfer of electrons onto positively charged donors is related to formation of the DX^- states, which localize two electrons becoming negatively charged.[9] Consequently, a situation when donors in GaAs and $Al_xGa_{1-x}As$ are partially occupied with electrons corresponds to the presence of positively (d^+) and negatively (DX^-) charged objects, both of which contribute to scattering. Due to electrostatic interactions, in thermal equilibrium conditions, the spatial arrangement of d^+ and DX^- states becomes correlated.[6,10–17] The spatial correlation of donor charges modifies significantly the carrier mobility. This results from the fact that d^+-DX^- dipoles represent much weaker carrier scattering efficiency compared with independent d^+ and DX^- states.

One can expect that the effects of correlations within the charged impurities might influence a transport mechanism of 2DEG.[18,19] A proper approach to determine the Coulomb electron scattering in such systems should take into account interference of single-remote-impurity potentials.[20,21] We will demonstrate that a significant contribution to an enhancement of μ and originating from spatial correlations of impurity charges can be identified in modulation-doped heterostructures. In our experiments we have found the increase of μ by a factor one-third of its value corresponding to a random distribution of remote-impurity charges. This finding is supported by the results of our model calculations.[16] Correlations in positions of impurity charges lead to a radical decrease of fluctuations of the potential. This in turn lowers the probability of scattering of 2DEG due to charged impurities. This effect may be included in the standard description of the scattering through the structure factor $S(q)$, describing the correlations of the positions of impurity charges. In the 3D case this approach has been successfully applied to

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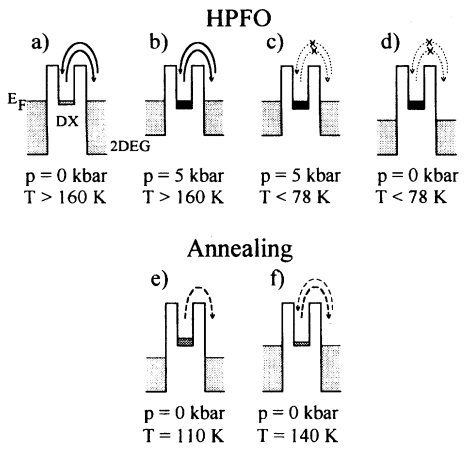

FIG. 1. Schematic illustration of the procedures leading to changes in the spatial distribution of donor centers, d^+ and DX^-. (a)–(d) represent high-pressure freeze-out of electrons onto DX^- states (HPFO) and (e) and (f) correspond to the annealing of samples at different temperatures. Note the solid and dotted arrows which stand for free and hindered transfer of electrons between the conduction band and DX^- system due to barriers for electron emission and capture. The dashed arrows correspond to partially transparent barriers.

GaAs:Si,[6,10,11] $Ga_{1-x}Al_xAs$:Si,[22] and HgSe:Fe.[6] In an idealized heterojunction model with donors distributed on a single plane at 10 nm distance from the 2D channel, inclusion of the correlation effects in the standard formula for the ionized impurity scattering[23] gives 3.5 times increase in the relaxation time for the correlated system, compared to random distribution of impurity charges. The correlations were obtained from Monte Carlo studies described in detail elsewhere.[24]

Experimental testing of the existence of the spatial-correlation-related contribution to the 2DEG transport consists of examining μ as a multivalued function of the 2DEG concentration n. We will demonstrate that depending on the method of the "preparation" of the same heterostructure, various values of μ can be obtained for the same density of 2DEG. This is interpreted as originating from various distributions of charges among the donor sites within the $Al_xGa_{1-x}As$ barrier. In our experiment we compare 2DEG mobilities (at $T=4.2$ K) measured for the most favorable spatial correlation of charges for a given amount of occupied donors (obtained during slow cooling of the sample) with a mobility value corresponding to the situation with reduced correlations.

To alter 2DEG density (i.e., an amount of occupied DX centers) and μ we employed two procedures [the first one, Figs. 1(a)–1(d), shows strong correlations; the second one, Figs. 1(e) and 1(f), shows weak correlations only]. Both procedures make use of the metastable properties of DX centers related to the existence of thermodynamic barriers for electron emission from and capture to the localized state of the donor. For Si donor the barriers block the electron transfer at temperatures below about 100–160 K.[8] This means that if a typical modulation-doped GaAs/$Al_xGa_{1-x}As$ heterostructure is cooled below this temperature range, a distribution of electrons on the impurity states and in the conduction band freezes.

(a) *High-pressure freeze-out*[11,13,15] (HPFO) of electrons on the metastable DX^- states. This procedure consists in applying hydrostatic pressure at temperatures above 160 K and releasing it at $T<100$ K. After applying hydrostatic pressure the donor levels corresponding to DX^- states penetrate deeper to the energy gap of $Al_xGa_{1-x}As$ which results in the enhanced transfer of electrons from the 2DEG to the donor states provided that the electron thermal energy exceeds the energy barrier [Fig. 1(b)]. At temperatures below about 100 K releasing the pressure does not change n because the trapped electrons cannot overpass the energy barrier between DX^- and the band states [Figs. 1(c) and 1(d)]. Providing spatial correlations in the distribution of these states, the HPFO procedure allows us to achieve the maximum value of μ for each value of n (determined by the magnitude of the freeze-out pressure). This effect was proved to reproduce μ_{max} versus n for bulk GaAs (Ref. 13) and bulk $Al_xGa_{1-x}As$.[15]

(b) *Annealing*. Employing HPFO procedure makes it possible to form a reservoir of localized electrons, which, at atmospheric pressure, are frozen in a metastable manner on DX^- states [Fig. 1(d)]. One can then use thermal excitation to transfer these electrons to the 2D channel. This consists of a subsequent annealing of the sample to temperatures above about 100 K and then cooling down to 4.2 K for performing measurements of n and μ. Each successive annealing step requires heating of the heterostructure to a higher temperature. Increasing temperature induces transfer of a portion of electrons from a metastable DX^- state to the 2D channel [Fig. 1(e)]. In the range of annealing temperatures between 110 and 140 K, changes in the impurity-charges distribution are caused by the electron emission from randomly "chosen" DX^- centers. At this temperature range retrapping of electrons onto DX^- centers and thus rearrangement of their spatial positions minimizing Coulomb interactions is hindered by capture barrier. This results in a reduction in the amount of the correlations.[24] A higher annealing temperature causes ionization of a higher amount of DX^- states to d^+ states as well as induction of correlations in spatial positions of impurity charges since a capture barrier becomes partially transparent [Fig. 1(f)]. In other words, during the annealing processes both the Fermi energy and electron kinetic energy increase leading to the decrease of the effective capture barrier. In addition, since the concentration of d^+ and 2D electrons raises significantly, the transfer of carriers from 2DEG to DX^- states is enhanced.

We have used two heterostructures with lower and higher values of μ (about 0.5 and 1.2×10^6 $cm^2/V\,s$ at $T=4.2$ K, respectively). Table I gives a description of the employed samples.

First, we will describe results of applying the HPFO procedure to the sample characterized by the lower value of μ (sample 1). Open circles on Fig. 2(a) represent the obtained results. Increasing freeze-out pressure during subsequent sample cooling-down processes (highest pressure 5 kbar) results in the decrease of both n and μ measured at $T=4.2$ K. An interplay between two effects determines the observed lowering of μ: a modification of the correlations and a decrease in the screening of scattering potential by 2DEG (lower concentration of carriers). We should emphasize here that processes of electron transfer between remote impurities

TABLE I. Parameters of the $GaAs/Al_xGa_{1-x}As$ modulation-doped heterostructures.

Sample	n (T=4.2 K) (10^{12} cm^{-2})	μ (T=4.2 K) (10^6 cm^2/V s)	Spacer (nm)	x (%)	Doping layer thickness (nm)	Doping layer density (10^{18} cm^{-3})
1	0.5	0.48	10	35	25	4
2	0.365	1.16	24	29	36	2.8

and 2DEG keeps the total number of charged scattering centers constant: $d^+ + 2e^- \leftrightarrow DX^-$.

The asterisks illustrate variation of μ with increasing n obtained after annealing of the sample. The increase of μ due to improvement of the screening was expected. In contrary, the first three cycles of the annealing (T is raised up to about 110 K) induce a decrease of μ. The origin of the μ reduction which accompanies the increase of n lies in a decrease in the amount of spatial correlations of d^+ and DX^- states. Thermal ionization of DX^- states occurs in a random manner. Consequently it leads to a more efficient 2DEG scattering due to remote impurities. Further annealing processes performed at higher temperatures cause a mobility increase although the "annealing branch" of the μ versus n dependence gives the mobility values lower than those achieved during HPFO. We must point out that within this temperature region the tendency to reduce the correlation in the arrangement of DX^- and d^+ states of Si donors competes with a process of equilibrium redistribution of electrons among the donor states. The latter mechanism leads to reestablishing of the correlations.

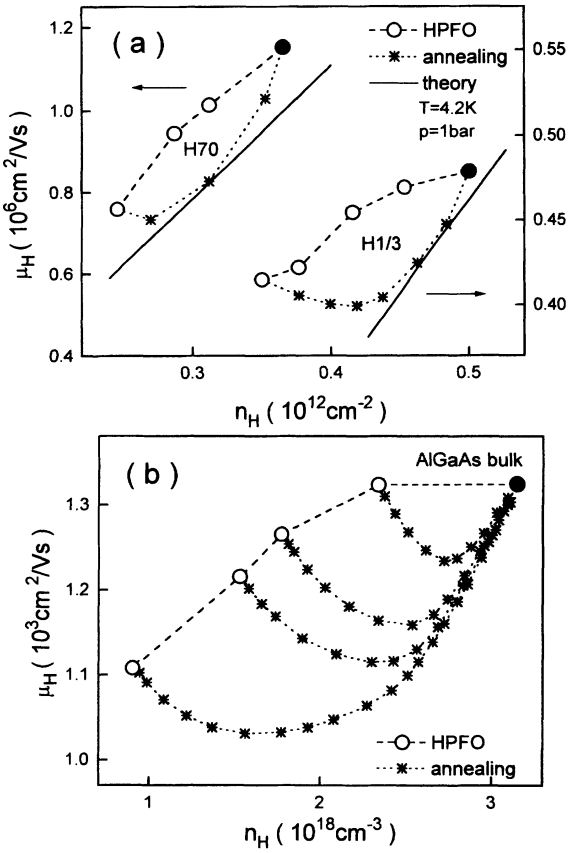

FIG. 2. (a) 2DEG mobility variation with electron concentration measured at T=4.2 K in samples 1 and 2. Open circles represent a strong correlation path achieved by HPFO procedure performed at different freeze-out pressures, whereas asterisks correspond to a weak correlation path produced by the subsequent sample annealing. Filled circles correspond to μ and n values obtained after cooling of the samples at ambient pressure. Solid lines represent results of theoretical calculations performed under the assumption of absence of the spatial correlations (Ref. 25). (b) 3D mobility vs n measured in "bulk" $Al_{0.15}Ga_{0.85}As$ sample and corresponding to $T \cong 78$ K. HPFO (○) and annealing (*) were used to modify occupation of Si-DX centers (Ref. 15). Size of symbols corresponds to the estimated experimental errors.

The solid lines in Fig. 2(a) represent μ versus n calculated under the assumption of absence of the spatial correlations.[25] Some information about the magnitude of the effect leading to mobility enhancement and originating in the spatial correlation of the donor charges can be deduced from a comparison of two μ values corresponding to the same 2DEG density. The maximum difference in the mobility values on the two branches of μ is about 15% which sets the lower limit of the examined effect.

To determine whether the correlation contribution to μ could be seen in a modulation-doped heterostructure with a larger spacer and higher mobility we have repeated the same sequence of measurements using sample 2. The results obtained [Fig. 2(a)] clearly show that the increase of μ for this sample reaches about 30% of its value for weakly correlated system.

To explain the origin of the multivalued function of μ versus n in modulation-doped heterostructures we have anticipated an existence of the spatial correlation of remote impurity charges in the barriers of studied samples. Are there other possible reasons for appearance of the observed effect? One can consider, for example, effects which resemble an anomalous behavior of μ due to conducting inhomogeneities in bulk, lightly doped GaAs.[26] Concerning our samples, it would mean that depending on the methods of their preparation, regions of higher and lower conductivity would be created (e.g., a parallel conduction in the region of barriers). In our opinion it should result in nonlinear dependence of the Hall voltage, u_{xy}, versus magnetic field or discrepancies in 2D electron gas concentration obtained from the Hall and Shubnikov–de Haas (SdH) effect measurements. We have performed measurements of u_{xy} and resistivity up to 14 T for sample 2. Linear changes of u_{xy} with the magnetic field (beside regions of the quantum Hall plateaus) and the same value of electron concentration deduced from Hall and SdH measurements seem to eliminate conducting inhomogeneities as a possible source of our findings.

The second situation we will discuss consists of an assumption that during sample annealing a higher number of negatively charged *DX* centers is produced in a close vicinity of a spacer than during applying HPFO procedure (for the same n value). Let us assume for a while that depending on the method of varying n different values of μ could appear. To examine this hypothesis we have decided to perform a similar comparison of μ versus n for the annealing and HPFO procedures in a 2-μm-thick layer of $Al_{0.15}Ga_{0.85}As$:Si grown on semi-insulating GaAs substrate with undoped $Al_{0.15}Ga_{0.85}As$ buffer layer. We have chosen a sample with low content of AlAs to achieve a condition (fulfilled in the heterostructures used) that almost all donors remain ionized during samples cooling at ambient pressure. Figure 2(b) shows the obtained results. μ represents again the multivalued function of n, which eliminates a requirement of the assumption that only preferential occupancy of more or less distant regions of doped barrier (with spatial correlations absent) explains the observed behavior of μ in $GaAs/Al_xGa_{1-x}As$ heterostructures.

In conclusion, we examined variation of 2DEG mobility with carrier concentration in different heterostructures of $GaAs/Al_xGa_{1-x}As$. We have shown that depending on the way the impurity charges are distributed among the donor sites in the $Al_xGa_{1-x}As$ barrier, different values of μ can be achieved for the same 2DEG density. The observed behavior of μ can be qualitatively attributed to changes in spatial correlation of the remote donor charges. This effect is responsible for a strong reduction of the 2DEG scattering by ionized remote impurities. This finding requires a qualitative modification of the widely accepted approach[27] to the electron scattering in modulation-doped heterostructure (as well as in doped superlattices, quantum wires, and quantum dots).

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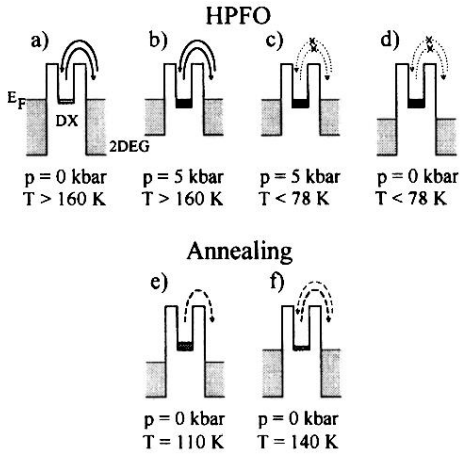

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