

Charge Carrier Dynamics in Ga_{1-x}Mn_xAs Studied by Resistance Noise Spectroscopy

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We report on electronic transport measurements of the magnetic semiconductor Ga_{1-x}Mn_xAs, whereby the defect landscape in various metallic thin films ($x = 6\%$) was tuned by He-ion irradiation. Changes in the distribution of activation energies, which strongly determine the low-frequency $1/f$ -type resistance noise characteristics, were observed after irradiation and can be explained by deep-level traps residing in the As sublattice. Various other kinds of crystalline defects such as, for instance, Mn interstitials, which possibly form nanoscale magnetic clusters with a fluctuating spin orientation, also contribute to the $1/f$ noise and can give rise to random telegraph signals, which were observed in films with $x = 7\%$. In addition, we neither find evidence for a magnetic polaron percolation nor any features in the noise near the Curie temperature.

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1. Introduction

Ga_{1-x}Mn_xAs, one of the first III-V diluted magnetic semiconductors [1], is considered a promising candidate for spintronics applications. Mn atoms substituted at Ga sites act both as acceptors and localized spins, between which a ferromagnetic order is mediated by holes below the Curie temperature T_C [2]. However, there is still no consensus on the nature of the electronic states as well as on the development of spontaneous magnetization [3, 4]. For samples with localized charge carriers, a percolation of bound magnetic polarons is an intriguing concept [5] in order to describe the origin of spontaneous magnetization. Resistance noise spectroscopy has proved itself as a powerful tool to detect a possible concomitant electronic phase separation [6]. Besides the investigation of possible electronic inhomogeneities in metallic Ga_{1-x}Mn_xAs films, we study the low-frequency carrier dynamics in order to probe the crucial role of defects in the present samples. In Ga_{1-x}Mn_xAs, a variety of electrically active defects exist – in particular Mn interstitials and As antisites. Both these defects act as double donors, hence compensating the hole doping and thereby suppressing ferromagnetic order. Previous noise studies suggest the formation of fluctuating nanoscale magnetic clusters [7], composed of Mn interstitials, which couple antiferromagnetically with substitutional Mn atoms. By He-ion irradiation we tune the carrier density without changing the Mn content x of the film. In detail, the main effect of ion

irradiation is the compensation of holes by introducing deep traps and thereby enhancing disorder, while both the concentration of substitutional Mn atoms and Mn interstitials remain unaltered [8]. A majority of the induced defects reside in the As sublattice [9].

2. Experimental details

We investigated three Ga_{1-x}Mn_xAs epitaxially grown thin film samples with $x = 6\%$ and a thickness of $d = 25$ nm, two of which had been irradiated with He-ions at HZDR (Dresden) using different doses, and two more films with $x = 7\%$ and $d = 25$ nm grown at PTB Braunschweig. The utilized ion beam energy was 4 keV and the corresponding fluences were 2.5×10^{13} cm⁻² and 3.5×10^{13} cm⁻² for the two irradiated samples. Samples were grown on semi-insulating GaAs(001). Further details regarding the sample growth and ion irradiation can be found in Ref. [8] and [10]. Curie temperatures for the $x = 6\%$ films, as determined from magnetization measurements, are 125 K (unirradiated sample), 75 K (low dose) and 50 K (high dose). Resistance and noise measurements were carried out in a four-terminal AC or DC setup. From noise measurements, we obtain the normalized power spectral density (PSD) $S_R = S_V/I^2$. Further details about the fluctuation spectroscopy can be found elsewhere [11]. In our experiments, external magnetic fields were applied perpendicular to the film plane.

3. Results and discussion

At first, we focus on the film ($x = 6\%$) irradiated with a low He-ion dose (fluence: 2.5×10^{13} cm⁻²), which shows a $1/f^\alpha$ frequency dependence of the noise in the entire temperature range. Here, α denotes the frequency exponent. Figure 1(a) shows a logarithmic plot of S_R/R^2 evaluated at 1 Hz against temperature T for different magnetic

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fields. Between 200 and 50 K we observe a decrease of the PSD by two orders of magnitude, followed by a drastic increase below 20 K. Similarly, the resistance $R(T)$, cf. inset of Fig. 1(a), exhibits a minimum followed by an-

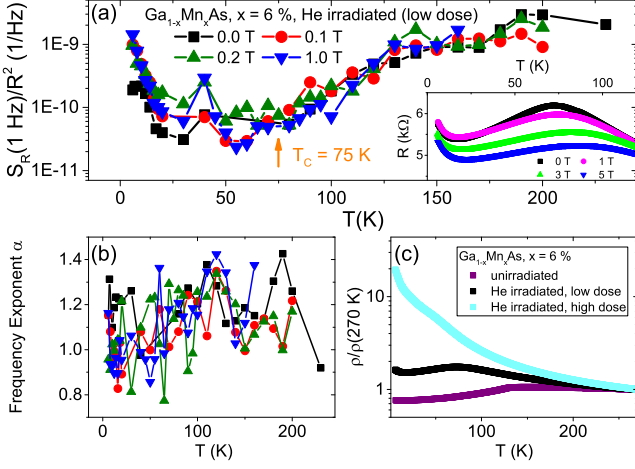


Fig. 1. (a) Logarithmic plot of S_R/R^2 at 1 Hz against T for the film ($x = 6\%$) irradiated with a low He-ion dose for different magnetic fields. Inset shows the resistance $R(T)$. (b) Frequency exponent α versus T as determined from noise spectra. (c) Resistivity $\rho(T)$ for all three samples with different irradiation doses normalized with regard to $\rho(T = 270 \text{ K})$.

noise magnitude in the hopping regime for doped semiconductors [12], which is in agreement with our observations. Furthermore, for all three $x = 6\%$ films, there is no change of S_R/R^2 in the presence of magnetic fields between 0 and 1 T and no occurrence of pronounced features around T_C . Figure 1(b) displays $\alpha(T)$ ranging from 0.8 to 1.4 within the entire temperature range and showing only a weak temperature dependence with no major changes in external magnetic fields. A comparison of the normalized resistivity $\rho/\rho(270 \text{ K})$ to the sample with higher irradiation dose (fluence: $3.5 \times 10^{13} \text{ cm}^{-2}$) and the unirradiated film is shown in Fig. 1(c), yielding a crossover from metallic towards more insulating behavior as a function of the utilized dose. In addition, as discussed in more detail in Ref. [10], S_R/R^2 also changes for different irradiation doses. Here, we present a further analysis of the $1/f$ -type noise and the underlying distribution of activation energies in order to learn about the nature of defects contributing to the $1/f$ noise. For this purpose, we apply the Dutta-Dimon-Horn (DDH) model [13], where a particular distribution of activation energies $D(E, T)$ determines the temperature dependence of both α and S_R/R^2 . A fundamental justification for the applicability of the DDH-model is the agreement of $\alpha(T)$ calculated after

$$\alpha(T) = 1 - \frac{1}{\ln(2\pi f \tau_0)} \left[\frac{\partial \ln\left(\frac{S_R}{R^2}(f, T)\right)}{\partial \ln(T)} - g'(T) - 1 \right] \quad (1)$$

with the experimentally determined values. Here, τ_0 denotes an attempt time, usually in the order of typical inverse phonon frequencies. Moreover, it is $g'(T) = \frac{\partial \ln(g(T))}{\partial \ln(T)} \equiv b$, where $g(T) = a \cdot T^b$ may account for an explicit temperature dependence of $D(E, T)$, which can occur due to a change of the number of thermally activated switching events or the coupling of fluctuations to the resistivity with temperature. We find a reasonable agreement for the unirradiated sample, cf. Fig. 2(a), and good agreement for the irradiated samples, cf. Fig. 2(b) and (c). The agreement between theory and experiment allows for calculating $D(E) \propto S_R/R^2 \times 2\pi f/k_B T \times 1/g(T)$ which is shown in Fig. 2(d)-(f), whereby $E = -k_B T \ln(2\pi f \tau_0)$. For each sample we observe a monotonous increase of $D(E)$ towards higher activation energies, which we interpret as a superposition of several thermally activated processes with different energies, that can be attributed to various kinds of defects. The film with a high irradiation dose shows a pronounced maximum at $E \approx 180 \text{ meV}$, which can presumably be explained by the high density of intentionally introduced defects due to the heavy irradiation with ions, causing trapping and detrapping processes of charge carriers with this particular energy. An alternative explanation is the spin-dependent scattering from weakly interacting nanoscale magnetic clusters with fluctuating spin orientation, which form after the irradiation process due to the increased disorder [9]. However, we do not find a systematic magnetic field dependence of the

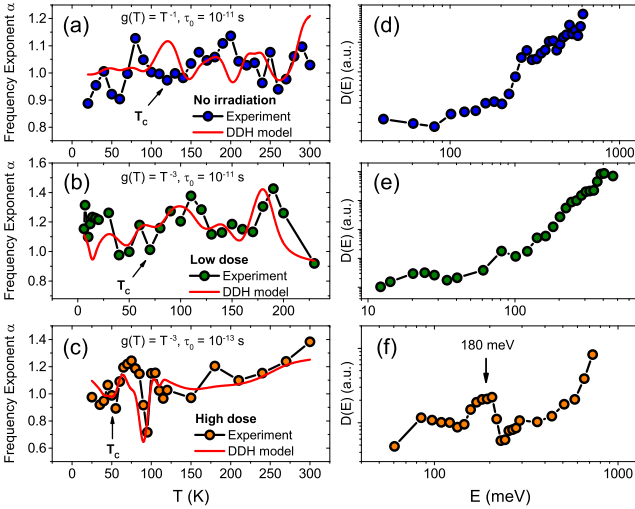


Fig. 2. (a)–(c) Comparison of the experimentally measured $\alpha(T)$ with predictions from the DDH-model for samples ($x = 6\%$) with different irradiation doses. (d)–(f) Distribution of activation energies $D(E)$ as determined from the DDH-model.

crease towards very low temperatures and can be fitted (not shown) by either the Mott variable range hopping (VRH) model, where $\ln(R) \propto T^{-1/4}$, or assuming a Kondo screening of magnetic moments by localized charge carriers, leading to $R \propto \ln(T)$. Both models parameterize the resistance behavior below 15 K equally well. In the case of Mott VRH, Burin et al. predict a steep increase of the

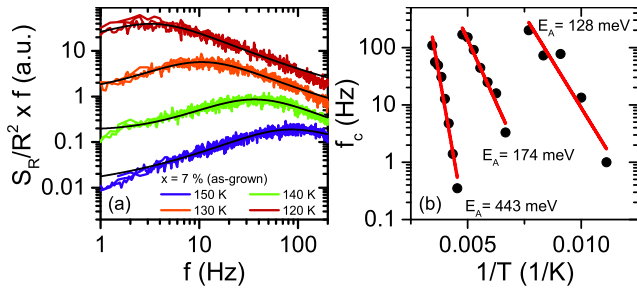


Fig. 3. (a) $S_R/R^2 \times f$ against f showing Lorentzian spectra for the $x = 7\%$ film (as-grown) for different T . For better overview, spectra were shifted. (b) Arrhenius plot of the corner frequency f_c against $1/T$. Linear fits yield the activation energies E_A of the underlying two-level fluctuation processes.

resistance noise. In general, $1/f$ -type noise is constituted of a superposition of many two-level fluctuation processes with different time constants, each represented by a Lorentzian spectrum. However, in certain cases, a dominating Lorentzian spectrum is superimposed on the $1/f$ -type signal, resulting in

$$\frac{S_R(f)}{R^2} \cdot f = \frac{C_1}{f^{\alpha-1}} + \frac{C_2}{4\pi^3} \cdot \frac{f}{f^2 + f_c^2}, \quad (2)$$

where C_1 and C_2 are constants denoting the magnitude of the $1/f$ -spectrum and the strength of the individual fluctuators, respectively. Furthermore, $f_c = 1/\tau_c$ represents the corner frequency. In the time domain, a Lorentzian spectrum corresponds to a random telegraph signal. Figure 3(a) shows exemplary spectra between 120 K and 150 K for the as-grown film with $x = 7\%$. Here, $S_R/R^2 \times f$ is plotted against f and the spectra are shifted with respect to each other for clarity. For individual thermally activated two-level processes, the corner frequency shifts as $f_c = f_0 \exp(-E_A/k_B T)$. Thus we can determine the activation energies E_A from an Arrhenius representation as shown in Fig. 3(b). In this case, linear fits yield values for E_A of about 128 meV, 174 meV and 443 meV. For measurements in external magnetic fields between $B = 0.1$ T and 3 T, we obtain similar activation energies around 100 meV and between 400 meV and 600 meV with no significant changes as a function of B . An additional sample with $x = 7\%$, treated by post-growth annealing at 200°C (18 hours in ambient atmosphere, cf. Ref. [10]), yields comparable results. The observation of Lorentzian spectra in the $x = 7\%$ films can be ascribed to a greater density of electrically active Mn interstitials due to the higher Mn flux rate utilized during the growth procedure, leading to an enhanced rate of capture and emission processes of charge carriers, since, for example, the activation energy of about 100 meV strongly resembles the energy level of substitutional Mn acceptors, which are situated 110 meV above the valence band edge. Alternatively, spin-dependent scattering from fluctuating nanoscale magnetic clusters is conceivable [7]. However, we detect two-level fluctuators in a much broader range $50 \text{ K} \leq T \leq 300 \text{ K}$ with higher activation energies than

in Ref. [7] and do not detect any magnetic field dependence, which speaks for a different mechanism causing the random telegraph signal.

4. Conclusions

In conclusion, we analyzed the noise characteristics for a series of metallic $\text{Ga}_{1-x}\text{Mn}_x\text{As}$ thin films ($x = 6\%$) with the DDH-model and found that deep-level traps, intentionally introduced by He-ion irradiation, lead to changes in the distribution of activation energies $D(E)$. While for the non-irradiated sample a monotonic increase in $D(E)$ can be observed, a high fluence of ions yields a distinct peak at 180 meV. This can be explained by capture and emission processes of charge carriers or alternatively by spin-dependent scattering from magnetic clusters with fluctuating spin orientation [9]. Samples with a higher Mn content ($x = 7\%$) exhibit dominating two-level processes superimposed on the $1/f$ -type signal, possibly due to a higher density of substitutional and interstitial Mn atoms. Finally, no signs of a percolative transition with coexisting phases were detected in any of the films, but instead the noise is clearly dominated by defect physics.

References

- [1] H. Ohno, *Science* **281**, 5379 (1998).
- [2] T. Jungwirth, J. Sinova, J. Mašek, J. Kučera, A.H. MacDonald, *Reviews of Modern Physics* **78**, 809 (2006).
- [3] T. Dietl, *Science* **287**, 1019 (2000).
- [4] M. Dobrowolska, K. Tivakornsasithorn, X. Liu, J. K. Furdyna, M. Berciu, K. M. Yu, W. Walukiewicz, *Nature Materials* **11**, 444 (2012).
- [5] A. Kaminski, S. D. Sarma, *Phys. Rev. Lett.* **88**, 247202 (2002).
- [6] P. Das, A. Amyan, J. Brandenburg, J. Mueller, P. Xiong, S. von Molnár, Z. Fisk, *Phys. Rev. B* **86**, 184425 (2012).
- [7] M. Zhu, X. Li, G. Xiang, N. Samarth, *Phys. Rev. B* **76**, 201201 (2007).
- [8] S. Zhou, L. Li, Y. Yuan, A.W. Rushforth, L. Chen, Y. Wang, R. Böttger, R. Heller, J. Zhao, K.W. Edmonds, R.P. Campion, B.L. Gallagher, C. Timm, M. Helm, *Phys. Rev. B* **94**, 075205 (2016).
- [9] E.H.C.P. Sinnecker, G.M. Penello, T.G. Rappoport, M.M. Sant'Anna, D.E.R. Souza, M.P. Pires, J.K. Furdyna, X. Liu, *Phys. Rev. B* **81**, 245203 (2010).
- [10] M. Lonsky, J. Teschabai-Oglu, K. Pierz, S. Sievers, H. W. Schumacher, Y. Yuan, R. Böttger, S. Zhou, J. Müller, *Phys. Rev. B* **97**, 054413 (2018).
- [11] J. Müller, *ChemPhysChem* **12**, 1222 (2011).
- [12] A.L. Burin, B.I. Shklovskii, V.I. Kozub, Y.M. Galperin, V. Vinokur, *Phys. Rev. B* **74**, 075205 (2006).
- [13] P. Dutta, P. Dimon, P.M. Horn, *Phys. Rev. Lett.* **43**, 646 (1979).