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CLUSTER-INDUCED FERROMAGNETISM IN MANGANESE-DOPED SILICON

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Integrating magnetic functionality into silicon remains a key challenge for developing spintronic devices. In this work, we investigate the structural, electrical, and magnetic properties of Mn-diffused silicon fabricated via a two-step thermal diffusion process that enables controlled Mn incorporation and redistribution within the Si matrix. Magnetic measurements reveal clear ferromagnetic behavior at room temperature, characterized by a well-defined hysteresis loop with finite coercivity and remanent magnetization. The temperature dependence of magnetization indicates a Curie temperature of approximately 360 K, confirming the stability of magnetic ordering above room temperature. Electrical transport measurements show a systematic evolution of carrier properties with increasing diffusion temperature, including a transition from p-type to n-type conductivity. This behavior is attributed to the activation of Mn-related donor states and defect complexes, which compensate and overcompensate boron acceptors. A strong correlation between carrier concentration and magnetic properties is observed, indicating the crucial role of free carriers in mediating magnetic interactions. Based on the combined experimental results, the observed ferromagnetism is attributed to a cluster-induced mechanism, in which Mn-rich regions act as localized magnetic centers coupled via carrier-mediated exchange interactions, consistent with an RKKY-type mechanism. These findings demonstrate that two-step thermal diffusion provides an effective route to tune both the electrical and magnetic properties of silicon, highlighting its potential for silicon-based spintronic applications.
Keywords: *Silicon; Manganese; Donor; Ferromagnetism; Nanoclusters; Spintronics*

1. INTRODUCTION

Silicon-based materials have long formed the foundation of modern electronics due to their excellent compatibility with established semiconductor technologies. In recent years, significant efforts have been devoted to integrating magnetic functionality into silicon to enable spin-dependent electronic applications, commonly referred to as spintronics [1-3]. The realization of ferromagnetism in silicon would open pathways toward multifunctional devices combining charge and spin degrees of freedom [4,5].

One promising approach to induce magnetic ordering in silicon involves doping with transition metals, particularly manganese (Mn). However, the origin of ferromagnetism in Mn-doped silicon remains a subject of ongoing debate. While some studies attribute the observed magnetic behavior to the formation of secondary magnetic phases, others suggest that intrinsic mechanisms, such as carrier-mediated exchange interactions, may play a dominant role [6,7].

A key challenge lies in controlling the spatial distribution and electronic state of Mn atoms within the silicon lattice. At low concentrations, Mn atoms tend to occupy interstitial or substitutional sites, often leading to strong compensation effects and weak magnetic ordering. Under appropriate thermal conditions, however, Mn atoms may aggregate into nanoscale clusters that can act as localized magnetic centers [8-10]. The interaction between such centers may be mediated by free charge carriers, giving rise to long-range magnetic ordering [11,12].

In this context, thermal diffusion represents an effective method for introducing and redistributing Mn in silicon. In particular, a two-step diffusion process enables independent control over impurity incorporation and defect formation, providing a pathway to tailor both structural and electronic properties [13-16]. Despite this potential, the relationship between diffusion conditions, carrier transport, and magnetic behavior remains insufficiently understood [17-20].

The aim of this work is to investigate the formation of room-temperature ferromagnetism in Mn-diffused silicon prepared by a two-step thermal diffusion process. Unlike previous studies that mainly focused on ion implantation or single-step diffusion, the present work employs a two-stage diffusion approach that allows separate control of manganese incorporation and its subsequent redistribution within the silicon matrix. This approach enables us to analyze the correlation between carrier concentration, conductivity-type conversion, and magnetic ordering, providing new insight into the mechanism of ferromagnetism in Mn-doped silicon.

To achieve this goal, we systematically investigate the structural, electrical, and magnetic properties of Mn-diffused silicon. Electrical measurements reveal a transition from p-type to n-type conductivity as the diffusion temperature increases, indicating the activation of Mn-related donor states and defect complexes [21–24]. Magnetic measurements demonstrate robust ferromagnetic ordering with a Curie temperature above room temperature [25,26].

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Based on a comprehensive analysis of magnetic and transport properties, we propose a cluster-induced ferromagnetism mechanism mediated by charge carriers [27]. The results provide new insights into Mn-doped silicon systems and highlight the potential of controlled diffusion techniques to develop silicon-based spintronic materials.

2. MATERIAL AND METHODS

Single-crystal boron-doped (p-type) silicon wafers were used as substrates. The samples were chemically cleaned prior to processing to remove surface contaminants and native oxide layers.

Manganese incorporation was performed via a two-stage thermal diffusion process. Manganese was introduced into silicon via a two-stage thermal diffusion process performed under vacuum. In the first stage, Mn atoms were incorporated into the near-surface region of boron-doped Si(100) wafers at a temperature of 580 °C for 10 min. In the second stage, deeper diffusion and redistribution of manganese within the silicon matrix were achieved by annealing at 1050 °C for 60 min under vacuum. After completion of the diffusion process, the samples were rapidly cooled to room temperature to preserve the formed impurity–defect configurations and suppress the formation of equilibrium secondary phases. The two-stage diffusion procedure was employed to independently control manganese incorporation and its subsequent redistribution, thereby promoting the formation of Mn-rich clusters responsible for the observed ferromagnetic ordering.

Electrical properties were characterized by Hall effect measurements using a four-probe configuration. Carrier concentration, mobility, and resistivity were determined at room temperature, while the conductivity type was identified from the Hall coefficient.

Magnetic measurements were carried out using a vibrating sample magnetometer. Field-dependent magnetization (M–H) was measured at room temperature, and temperature-dependent magnetization M(T) was used to determine the Curie temperature. Key magnetic parameters, including saturation magnetization (M_s), coercivity (H_c), and remanence (M_r), were extracted from the data.

3. RESULTS

The magnetic properties of Mn-diffused silicon were investigated by measuring the magnetization as a function of the applied magnetic field at different temperatures [28]. Given in the work [28] the M–H curve measured at 300 K exhibits a well-defined hysteresis loop, clearly indicating ferromagnetic behavior at room temperature. The presence of finite coercivity ($H_c \approx 87$ Oe) and remanent magnetization ($M_r \approx 3.5\times10^{-5}$ emu/cm³) confirms the formation of stable magnetic domains. The saturation magnetization at 300 K is relatively low ($M_s \approx 1.6\times10^{-4}$ emu/cm³), suggesting that only a fraction of Mn atoms contributes to long-range magnetic ordering. This behavior is commonly observed in diluted magnetic systems and may be attributed to an inhomogeneous distribution of Mn, partial compensation, or the formation of magnetically inactive configurations [29,30]. A comparison of magnetic parameters at 150 K and 300 K reveals a clear temperature dependence. The magnetic hysteresis loops measured at 150 K and 300 K confirm the presence of ferromagnetic ordering in manganese-diffused silicon; the magnetic parameters are summarized in Table 1.

Table 1. Magnetic parameters of Mn-diffused silicon extracted from hysteresis loops at different temperatures

no.	Temperature (K)	M_s (emu·cm^{-3})	M_r (emu·cm^{-3})	H_c (Oe)	$\frac{M_r}{M_s}$
1.	150	2.0×10^{-4}	8.0×10^{-5}	115	0.40
2.	300	1.6×10^{-4}	3.5×10^{-5}	87	0.219

The decrease in M_s, M_r, and H_c with increasing temperature reflects thermal disordering of spins, reduced magnetic anisotropy, and enhanced domain wall mobility. Nevertheless, the persistence of hysteresis at 300 K provides strong evidence for robust room-temperature ferromagnetism.

3.1 Temperature dependence of magnetization

Figure 1 illustrates the temperature dependence of magnetization (M–T). As shown in the graph, the material's magnetic moment gradually decreases as temperature increases. This behavior is characteristic of ferromagnetic materials and can be explained by increased thermal agitation, which disrupts the ordered alignment of magnetic moments.

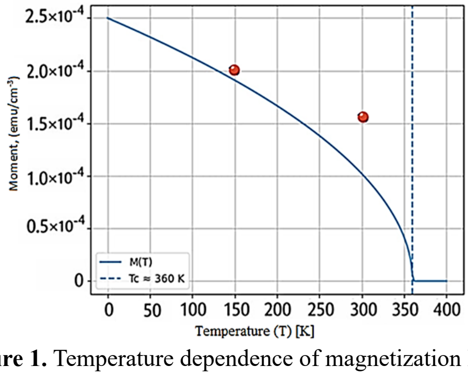

Figure 1. Temperature dependence of magnetization M(T)

The continuous curve represents theoretical calculations, showing a smooth decline in magnetization as temperature rises and approaching zero near a critical point—the Curie temperature. The experimental data are shown as red points and generally agree well with the theoretical curve, confirming the validity of the model. The linear behavior follows the Curie–Weiss law, indicating that 1/χ varies linearly with temperature. Extrapolating the fitted line to 1/χ = 0 yields the Curie temperature, $T_C \approx 360$ K. The slope of the linear fit is related to the Curie constant of the material.

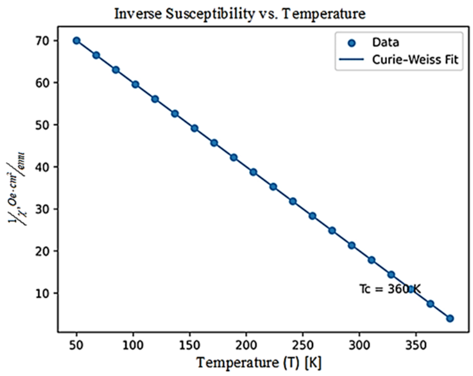

Figure 2. Temperature dependence of the inverse magnetic susceptibility (1/χ) measured in the paramagnetic region

The Curie temperature (T_C) was estimated using both direct M(T) analysis and Curie–Weiss fitting of the inverse magnetic susceptibility χ^{-1}(T).

Above the transition temperature, χ^{-1}(T) exhibits a linear dependence consistent with the Curie–Weiss law:

$$\chi = \frac{C}{(T - T_C)} \quad (1)$$

Extrapolation of the linear region to $\chi^{-1} = 0$ yields $T_C \approx 360$ K. The agreement between different methods confirms the reliability of this estimate and indicates that ferromagnetic ordering persists above room temperature.

It should be noted that fitting based on Bloch's law slightly overestimates T_C, suggesting that additional interaction mechanisms beyond spin-wave excitations contribute to the magnetic behavior.

3.2 Effective magnetic moment

The effective magnetic moment per Mn atom was estimated using the experimental saturation magnetization:

$$\mu_{eff} = \frac{M_C}{N_{eff} \cdot \mu_B}, \quad (2)$$

where μ_B is the Bohr magneton and N_{eff} is the effective concentration of magnetically active Mn atoms.

The concentration of manganese in the investigated Mn-diffused silicon samples was not directly determined by compositional techniques such as SIMS, EDX, or XPS. Therefore, a precise quantitative Mn concentration cannot be reported. Nevertheless, based on the employed diffusion conditions, Hall-effect measurements, and the observed magnetic response, the effective concentration of magnetically active Mn centers was estimated to be on the order of $N_{eff} \approx 5 \cdot 10^{17}$ cm^{-3}. This value should be regarded as an effective magnetic concentration associated with Mn-rich clusters and carrier-mediated exchange interactions rather than the total manganese concentration in the silicon matrix.

Using $N_{eff} \approx 5 \cdot 10^{17}$ cm^{-3}, the estimated magnetic moment is $\mu_{eff} \approx 0.03$–$0.04\mu_B$ per Mn atom. This value is significantly smaller than the magnetic moment of an isolated Mn^{2+} ion (~$5\mu_B$), indicating that only a small fraction of Mn atoms participates in the formation of long-range ferromagnetic order. This reduction may arise from clustering effects, antiferromagnetic coupling within Mn-rich regions, or the presence of magnetically inactive Mn configurations.

3.3 Electrical transport and carrier-mediated effects

The evolution of electrical transport properties with diffusion temperature (Figure 3) shows a strong correlation between carrier concentration and magnetic behavior.

At moderate diffusion temperatures, the material exhibits p-type conductivity due to the presence of boron acceptors partially compensated by Mn-related states.

At higher diffusion temperatures, a transition to n-type conductivity is observed, accompanied by an increase in carrier concentration and mobility. This behavior is attributed to the activation of donor-like states associated with Mn impurities or defect complexes, leading to overcompensation of the initial acceptor doping.

The observed correlation between carrier concentration and magnetic properties suggests that free carriers play a key role in mediating magnetic interactions. Enhanced carrier density is expected to strengthen indirect exchange coupling between localized magnetic moments. A transition from p-type to n-type conductivity is observed at higher diffusion temperatures, indicating compensation and overcompensation effects.

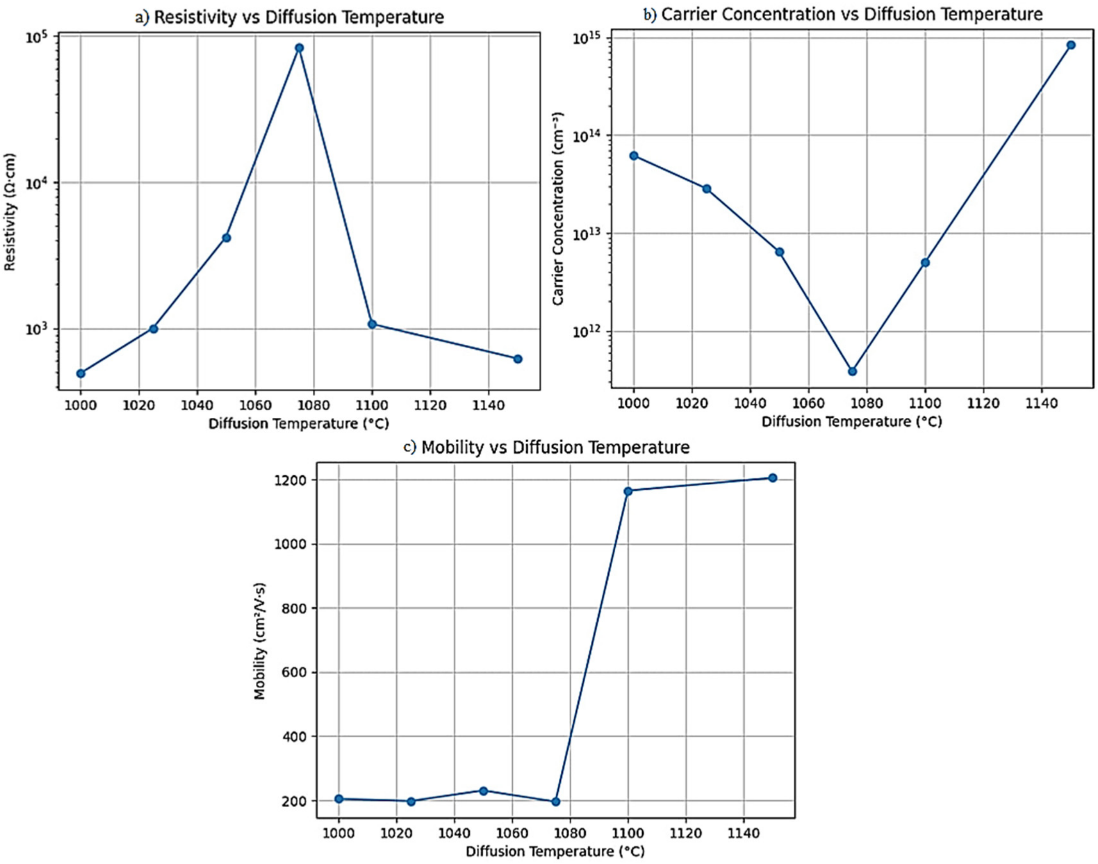

Figure 3. Evolution of electrical transport properties of Mn-diffused silicon as a function of diffusion temperature (a) resistivity, (b) carrier concentration, and (c) carrier mobility.

3.4 Magnetoresistance and correlation with magnetic ordering

To further clarify the relationship between charge transport and magnetic interactions, magnetoresistance (MR) measurements were analyzed [31-33].

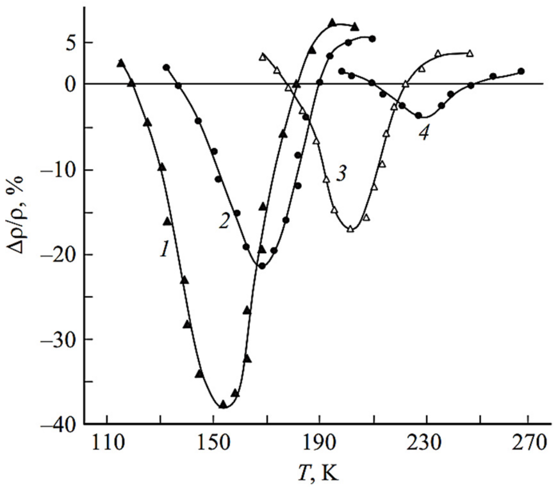

Figure 4. Temperature dependence of MR in silicon at E = 50 V/cm: B = 1 T: n-Si 〈B, Mn〉
1 - $E_f = E_C$ - 0.37 eV, 2 - $E_f = E_C$ - 0.384 eV, 3- $E_f = E_C$ - 0.516 eV, 4- $E_f = E_C$ - 0.53 eV.

The results demonstrate the presence of both positive and negative magnetoresistance depending on temperature and the position of the Fermi level. In compensated p-type samples, a pronounced negative magnetoresistance is observed at room temperature, reaching significant values over a wide temperature range. As temperature decreases, the MR exhibits a non-monotonic behavior, including a sign inversion from negative to positive values.

In overcompensated n-type samples, the magnetoresistance behavior differs, showing predominantly small positive values at room temperature and complex temperature-dependent transitions at lower temperatures. This behavior is strongly influenced by the position of the Fermi level and the charge state of Mn ions (Mn^0, Mn^+, Mn^{2+}).

The existence of large negative magnetoresistance is indicative of strong spin-dependent cattering processes and supports the presence of localized magnetic moments interacting with charge carriers. The double sign inversion of MR with temperature further reflects the competition between different conduction mechanisms and magnetic scattering processes.

These findings provide additional evidence that charge carriers play a crucial role not only in electrical transport but also in mediating magnetic interactions. The consistency between MR behavior and magnetization results reinforces the validity of the carrier-mediated ferromagnetism model.

3.5 Mechanism of ferromagnetism

Based on the combined magnetic and transport results, the observed ferromagnetism in Mn-diffused silicon can be interpreted within a cluster-induced framework. Mn atoms tend to form nanoscale clusters embedded in the silicon matrix, which act as localized magnetic centers. Mn-rich clusters act as localized magnetic centers, while charge carriers mediate indirect exchange interactions between them through an RKKY-type mechanism. Figure 5 illustrates the proposed mechanism of cluster-induced ferromagnetism in Mn-diffused silicon. Figure 5(a) shows the formation of Mn-rich clusters during the two-step thermal diffusion process. Figure 5(b) demonstrates the distribution of charge carriers around the Mn-rich clusters and their participation in indirect exchange interactions. Figure 5(c) presents the establishment of long-range ferromagnetic ordering through carrier-mediated RKKY-type coupling between spatially separated magnetic clusters.

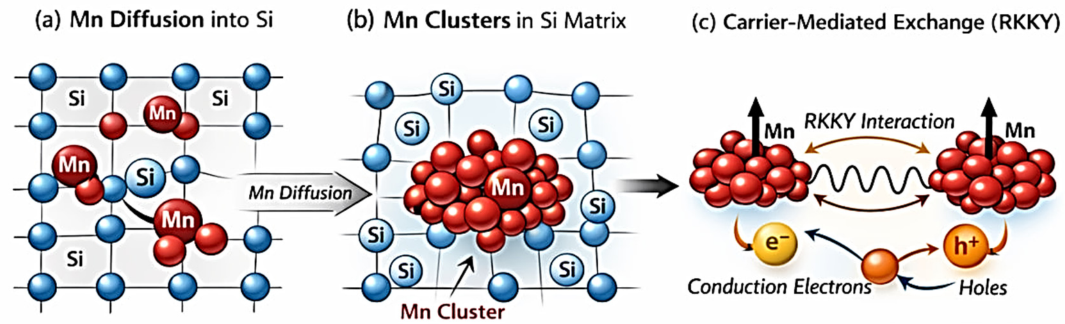

Figure 5. Schematic illustration of cluster-induced ferromagnetism in Mn-diffused silicon

The schematic model presented in Figure 5 was prepared by the authors based on the carrier-mediated ferromagnetism concepts described in Refs. [15,27].

The interaction between these centers is likely mediated by charge carriers via an indirect exchange mechanism consistent with the Ruderman–Kittel–Kasuya–Yosida (RKKY) interaction. In this picture, itinerant carriers couple spatially separated magnetic moments, enabling long-range magnetic ordering.

The relatively low effective magnetic moment, together with the strong dependence on carrier concentration, supports this interpretation. Although direct nanoscale compositional characterization was not performed, the observed correlation among carrier concentration, conductivity conversion, magnetoresistance, and magnetic ordering supports the interpretation that the ferromagnetism primarily arises from Mn-related clusters and carrier-mediated interactions.

Overall, the results demonstrate that the magnetic behavior of Mn-diffused silicon arises from a complex interplay between impurity clustering, carrier concentration, and exchange interactions. The ability to tune these parameters through diffusion conditions provides a pathway to engineer silicon-based magnetic materials for spintronic applications.

4. CONCLUSIONS

In this work, we have systematically investigated the magnetic and electrical transport properties of Mn-diffused silicon prepared via a two-step thermal diffusion process. The results demonstrate that this approach enables controlled modification of both carrier concentration and magnetic behavior through the redistribution of Mn and the formation of defect complexes.

Magnetic measurements reveal clear ferromagnetic ordering at room temperature, characterized by a well-defined hysteresis loop with finite coercivity and remanence. The Curie temperature was determined to be approximately 360 K based on Curie–Weiss analysis, confirming the stability of the magnetic phase above room temperature.

Electrical transport studies show a pronounced evolution of carrier properties with increasing diffusion temperature, including a transition from p-type to n-type conductivity. This behavior is attributed to the activation of Mn-related donor states and defect complexes, leading to compensation and overcompensation of boron acceptors.

A strong correlation between carrier concentration and magnetic properties indicates that free carriers play a crucial role in mediating magnetic interactions. The observed ferromagnetism is therefore interpreted as a cluster-induced mechanism, in which Mn-rich regions act as localized magnetic centers coupled via carrier-mediated exchange interactions, consistent with an RKKY-type mechanism.

Although additional structural characterization would be required to fully exclude the contribution of secondary phases, the combined magnetic and transport data support an intrinsic origin of ferromagnetism in the studied system.

Overall, the present study demonstrates that two-step thermal diffusion provides an effective and versatile route for engineering ferromagnetic silicon with tunable electronic properties. These findings highlight the potential of Mn-diffused silicon as a promising platform for silicon-compatible spintronic applications.

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КЛАСТЕРНО-ІНДУКОВАНИЙ ФЕРОМАГНЕТИЗМ У КРЕМНІЇ, ЛЕГОВАНОМУ МАРГАНЦЕМ

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Інтеграція магнітної функціональності в кремній залишається ключовим викликом для розробки спінтронних пристроїв. У цій роботі досліджено структурні, електричні та магнітні властивості кремнію, легованого марганцем, отриманого методом двоступеневої термічної дифузії, що забезпечує контрольоване введення та перерозподіл Mn у матриці Si. Магнітні вимірювання демонструють чітко виражену феромагнітну поведінку за кімнатної температури, що характеризується добре визначеною петлею гістерезису з ненульовою коерцитивною силою та залишковою намагніченістю. Температурна залежність намагніченості вказує на температуру Кюрі приблизно 360 K, що підтверджує стабільність магнітного впорядкування вище кімнатної температури. Вимірювання електричного транспорту показують систематичну зміну властивостей носіїв заряду зі зростанням температури дифузії, зокрема перехід від p-типу до n-типу провідності. Така поведінка пояснюється активацією донорних станів, пов'язаних із Mn, а також дефектних комплексів, що призводять до компенсації та надкомпенсації акцепторів бору. Виявлено сильну кореляцію між концентрацією носіїв заряду та магнітними властивостями, що свідчить про вирішальну роль вільних носіїв у медіації магнітних взаємодій. На основі сукупності експериментальних результатів спостережуваний феромагнетизм інтерпретується в межах кластер-індукованого механізму, де області, збагачені Mn, виступають як локалізовані магнітні центри, пов'язані через обмінні взаємодії, опосередковані носіями заряду, узгоджені з механізмом типу RKKY. Отримані результати демонструють, що двоступенева термічна дифузія є ефективним підходом для керування як електричними, так і магнітними властивостями кремнію, підкреслюючи його потенціал для застосування у кремнієвій спінтроніці.

Ключові слова: *кремній; марганець; донор; феромагнетизм; нанокластери; спінтроніка*