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Clustering effects of spin radicals on superparamagnetism: calculation for diamond with radiation-induced defects

N A Poklonski[1,*], S A Vyrko[1] and A G Zabrodskii[2]

[1] Physics Department, Belarusian State University, Nezavisimosti Ave. 4, Minsk 220030, Belarus
[2] Ioffe Institute, Politekhnicheskaya Str. 26, St. Petersburg, 194021, Russia
* Author to whom any correspondence should be addressed.

E-mail: poklonski@bsu.by

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Abstract

The influence of the clustering degree of electron spin magnetic moments from crystalline defects on the low-temperature magnetic susceptibility of materials is theoretically studied. The susceptibility of two diamond samples containing only identical spin radicals (uncompensated electron spins of radiation defects) with a concentration of 10^{19} cm^{-3} is calculated. The first sample contains only isolated spin radicals. The second sample contains the same spin radicals grouped into identical spherical clusters—ferrons (clusters). With an average sample concentration of 10^{19} cm^{-3}, the local concentration of spin radicals inside each ferron is assumed to be at the limiting value of 10^{20} cm^{-3}. As the number of spin radicals in each ferron increased from 10 to 350, the diameters of the ferrons increased from 6 to 19 nm, respectively. The low-temperature magnetic susceptibility of both samples is calculated based on the Brillouin function. It is shown that as the number of spin radicals in each ferron increases, the superparamagnetic behavior of the second sample is enhanced. At helium temperatures, in the limit of zero external magnetic field induction, the magnetic susceptibility of the second sample, in which the spin radicals are grouped into ferrons, is much greater than the susceptibility of the first sample, which contains only isolated spin radicals. This finding is supported by experimental measurements of low-temperature magnetic susceptibility in natural type IIa diamond samples irradiated with reactor neutrons.

1. Introduction

Imparting magnetic properties to non-magnetic semiconductor materials and enhancing these characteristics is crucial for expanding the range of functional materials suitable for magnetoelectronics [1, 2]. In this context, mono- and polycrystalline synthetic diamond films grown by chemical vapor deposition (CVD) show significant promise for spintronics and photonics applications [3, 4].

Clusters (aggregates) of point defects of the crystal structure were observed by electron microscopy in neutron-irradiated monocrystalline CVD diamonds [5]. Ferromagnetic states in a 20-nm-thick layer were detected at room temperature in laser-processed diamonds [6]. Vibrating-sample magnetometry of natural type IIa diamond grains irradiated with a neutron fluence of $\approx 5 \cdot 10^{20}$ cm^{-2} revealed ferromagnetic ordering of uncompensated electron spins of radiation defects in the 2–100 K temperature range [7]. During magnetization reversal of diamond grains with radiation defects at 2–4 K, a non-monotonic dependence of the differential magnetic susceptibility on the external magnetic field was observed, attributed to the magnetocaloric effect (cf. experiments with heavily doped germanium and silicon carbide crystals [8, 9]).

Thus, the problem of mathematical and computer modeling of the observed enhancement of magnetic activity in semiconductor materials—specifically enhancement driven by superparamagnetism unrelated to magnetic impurities, as observed in diamond—is highly relevant both fundamentally and practically. The aim of this work is to theoretically examine the influence of localized spin-radical clustering processes on diamond

magnetization. Calculation results are directly applied to experimental data interpretation of magnetic susceptibility in natural diamond partially disordered by reactor neutron irradiation.

In the following, we use SI units throughout the paper.

2. Basic relationships

2.1. Gas of isolated localized spin radicals

Let us consider the first sample containing a gas of non-interacting (isolated) spin radicals.

The total electron spin magnetic moment per unit volume of diamond for all isolated (non-interacting) localized radicals with spin 1/2 in an external stationary magnetic field is given by the Brillouin formula [10, 11]:

$$M_{\mathrm{r}} = \frac{N_{\mathrm{r}} g_{\mathrm{r}} \mu_{\mathrm{B}}}{2} \mathcal{B}_{1/2}\left(\frac{g_{\mathrm{r}} \mu_{\mathrm{B}} B}{2 k_{\mathrm{B}} T}\right), \tag{1}$$

where N_{r} is the volume (three-dimensional) concentration of spin radicals, 1/2 is the quantum number of the electron spin angular momentum of the radical, $g_{\mathrm{r}} = 2.0027$ is the g-factor of the electron not involved in covalent bonds with carbon atoms of the diamond matrix [12, 13], μ_{B} is the Bohr magneton, $B = \mu_0 H$ is the induction of the constant magnetic field, μ_0 is the magnetic constant, H is the magnetic field strength, k_{B} is the Boltzmann constant, T is the absolute temperature, and $\mathcal{B}_{1/2}(x)$ is the Brillouin function:

$$\mathcal{B}_{1/2}(x) = 2\coth(2x) - \coth(x) = \tanh(x); \qquad x = x_{1/2} = \frac{\mu B}{k_{\mathrm{B}} T} = \frac{g_{\mathrm{r}} \mu_{\mathrm{B}} B}{2 k_{\mathrm{B}} T}, \tag{2}$$

here $\mu = g_{\mathrm{r}} \mu_{\mathrm{B}}/2$ is the magnetic moment of a single spin radical; $\mathcal{B}_{1/2} = 0$ when $B = 0$.

For spin 1/2, the Brillouin function is $\mathcal{B}_{1/2}(x) = \tanh(x)$. From equation (1) taking into account equation (2), it follows that

$$M_{\mathrm{r}} = \frac{N_{\mathrm{r}} g_{\mathrm{r}} \mu_{\mathrm{B}}}{2} \tanh\left(\frac{g_{\mathrm{r}} \mu_{\mathrm{B}} B}{2 k_{\mathrm{B}} T}\right), \tag{3}$$

where $g_{\mathrm{r}} \mu_{\mathrm{B}}/2$ is the magnitude of the uncompensated magnetic moment of the radical electron with spin 1/2.

Differential magnetic susceptibility of a 'gas' of paramagnetic spin radicals in a diamagnetic matrix:[3]

$$\chi_{\mathrm{r}} = \frac{\mathrm{d}M_{\mathrm{r}}}{\mathrm{d}H}, \tag{4}$$

where M_{r} is the magnetization (magnetic moment per unit volume) of the sample due to radicals, and $H = B/\mu_0$ is the strength of the external magnetic field; the calculations do not take into account the dependence of the sample temperature on the magnetic field.

Substituting equation (3) into equation (4), for spin 1/2 we obtain:

$$\chi_{\mathrm{r}} = \frac{\mathrm{d}M_{\mathrm{r}}}{\mathrm{d}H} = \frac{\mathrm{d}M_{\mathrm{r}} \mu_0}{\mathrm{d}B} = \frac{N_{\mathrm{r}} (g_{\mathrm{r}} \mu_{\mathrm{B}})^2 \mu_0}{4 k_{\mathrm{B}} T} \frac{\mathrm{d}\mathcal{B}_{1/2}(x)}{\mathrm{d}x} \equiv \frac{N_{\mathrm{r}} (g_{\mathrm{r}} \mu_{\mathrm{B}})^2 \mu_0}{4 k_{\mathrm{B}} T} \mathcal{B}'_{1/2}\left(\frac{g_{\mathrm{r}} \mu_{\mathrm{B}} B}{2 k_{\mathrm{B}} T}\right), \tag{5}$$

where the first derivative of Brillouin function on magnetic field induction $\mathcal{B}'_{1/2}(x)$ has the form:

$$\mathcal{B}'_{1/2}(x) = \frac{\mathrm{d}\mathcal{B}_{1/2}(x)}{\mathrm{d}x} = \operatorname{csch}^2(x) - 4\operatorname{csch}^2(2x) = \operatorname{sech}^2(x); \qquad x = x_{1/2} = \frac{g_{\mathrm{r}} \mu_{\mathrm{B}} B}{2 k_{\mathrm{B}} T}. \tag{6}$$

2.2. Gas of ferrons (clusters of spin radicals)

Let us consider the second sample containing a gas of non-interacting (isolated) ferrons. The average concentration of spin radicals N_{r} over the sample is the same as in the first sample.

A ferron is a compact accumulation of uncompensated electron spins of radicals bound by ferromagnetic exchange in the form of a cluster, which can be considered as an analogue of a superparamagnetic nanoparticle with a sufficiently large magnetic moment, introduced by Néel [17] (see also [18, 19]).

The magnetic moment per unit volume (magnetization) of a diamond sample containing N_{fr} ferrons (neglecting their demagnetizing factor) can be expressed similarly to equation (1) as follows:

[3] The dimensions of magnetization M and magnetic susceptibility χ in different systems of units of physical quantities are as follows [14–16]: for SI: $[M]$ = A/m, $[\chi] = 1$ SI units; for CGS: $[M] = \mathrm{erg/(G{\cdot}cm^3)} = \mathrm{emu/cm^3}$, $[\chi] = \mathrm{emu{\cdot}cm^{-3}/Oe} = 1$ CGS units. The relationship between CGS and SI units for M and χ, respectively: 1 $\mathrm{emu/cm^3} = 10^3$ A/m; 1 CGS units = 4π SI units; $M_{\mathrm{SI}} = 10^3 M_{\mathrm{CGS}}$; $\chi_{\mathrm{SI}} = 4\pi \chi_{\mathrm{CGS}}$.

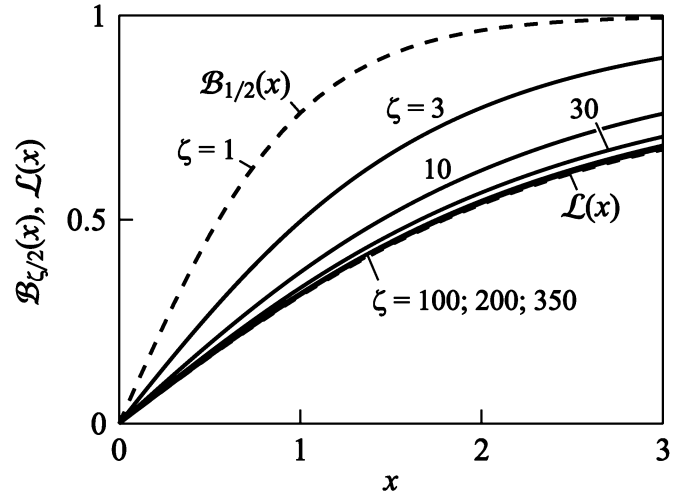

Figure 1. Behavior of the Brillouin function $\mathcal{B}_{\zeta/2}(x)$ for the number of uncompensated electron spins of radicals per ferron $\zeta = 1, 3, 10, 30, 100, 200, 350$ and the Langevin function $\mathcal{L}(x)$ depending on the parameter $x = \zeta g_r \mu_B B/2k_B T$.

$$M_{fr} = \frac{N_{fr}\zeta g_r \mu_B}{2}\mathcal{B}_{\zeta/2}\left(\frac{\zeta g_r \mu_B B}{2k_B T}\right), \tag{7}$$

where $N_{fr} = N_r/\zeta$ is the volume (three-dimensional) concentration of ferrons, each containing ζ radicals with uncompensated spins 1/2; $g_r = 2.0027$ is the g-factor of the spin-radical electron with spin 1/2 in the ferron (in the absence of orbital angular momentum for both single radicals [20] and their clusters [21]), and $\mathcal{B}_{\zeta/2}(x)$ is the Brillouin function for effective spin $\zeta/2$ of a single ferron:

$$\mathcal{B}_{\zeta/2}(x) = \frac{\zeta+1}{\zeta}\coth\left(\frac{\zeta+1}{\zeta}x\right) - \frac{1}{\zeta}\coth\left(\frac{1}{\zeta}x\right); \qquad x = x_{\zeta/2} = \frac{\zeta g_r \mu_B B}{2k_B T}. \tag{8}$$

When $\zeta = 1$, equation (7) reduces to equation (1) with $N_{fr} = N_r$.

Figure 1 illustrates the behavior of the Brillouin function for various ζ values and the Langevin function depending on the parameter x. As can be seen,[4] in the limit of the number of spin radicals per ferron $\zeta \to \infty$, the Brillouin function $\mathcal{B}_{\zeta/2}(x)$ for a single ferron with magnetic moment $\zeta g_r \mu_B/2$ according to equation (8) converges to the Langevin function $\mathcal{L}(x) = \coth(x) - 1/x$.

The differential magnetic susceptibility χ_{fr} of the ferron gas:

$$\chi_{fr} = \frac{dM_{fr}}{dH}. \tag{9}$$

Using equations (9) and (7) we derive

$$\chi_{fr}(\zeta) = \frac{dM_{fr}}{dH} = \frac{dM_{fr}\mu_0}{dB} = \frac{N_{fr}(\zeta g_r \mu_B)^2 \mu_0}{4k_B T}\frac{d\mathcal{B}_{\zeta/2}(x)}{dx} \equiv \frac{N_{fr}(\zeta g_r \mu_B)^2 \mu_0}{4k_B T}\mathcal{B}'_{\zeta/2}\left(\frac{\zeta g_r \mu_B B}{2k_B T}\right), \tag{10}$$

where the first derivative of the Brillouin function $\mathcal{B}'_{\zeta/2}(x)$ is

$$\mathcal{B}'_{\zeta/2}(x) = \frac{d\mathcal{B}_{\zeta/2}(x)}{dx} = \frac{1}{\zeta^2}\mathrm{csch}^2\left(\frac{1}{\zeta}x\right) - \left(\frac{1+\zeta}{\zeta}\right)^2\mathrm{csch}^2\left(\frac{1+\zeta}{\zeta}x\right); \qquad x = x_{\zeta/2} = \frac{\zeta g_r \mu_B B}{2k_B T}. \tag{11}$$

For $\zeta = 1$, equations (8) and (11) reduce to equation (6).

Let us separately consider the simplest case of extremely weak magnetic fields ($B \to 0$). In this limit, the Brillouin function according to equation (8) and its first derivative take the following form [23]:

$$\mathcal{B}_{\zeta/2}(x) = \frac{2+\zeta}{3\zeta}x, \quad \mathcal{B}'_{\zeta/2} = \frac{2+\zeta}{3\zeta}. \tag{12}$$

For $\zeta = 1$, from equation (12) we obtain that $\mathcal{B}'_{1/2} = 1$. Taking into account equation (12) for $B \to 0$, the magnetic susceptibility by equation (10) takes the form:

[4] The plots shown in figure 1 coincide with theoretical calculations of Brillouin and Langevin functions from [11, 22] when ζ is replaced by $2J$, where $J = L \pm S$ is the quantum number of the total angular momentum of the atomic electrons (Hund's rule), L is the quantum number of the orbital angular momentum, and S is the quantum number of the spin angular momentum.

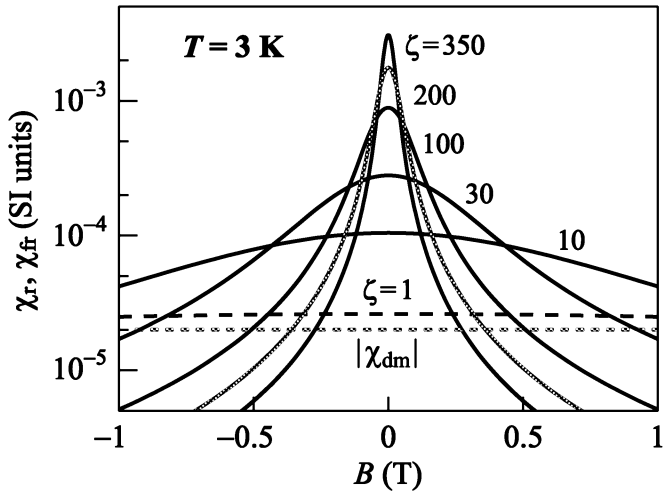

Figure 2. Differential magnetic susceptibility of isolated spin radicals $\chi_r \equiv \chi_{fr}$ for $\zeta = 1$ by equation (5) and ferrons (clusters of these radicals) $\chi_{fr}(\zeta)$ by equation (10) at temperature $T = 3$ K, $\zeta = 10, 30, 100, 200, 350$, and concentration of radicals $N_r = 10^{19}$ cm^{-3} as a function of the induction of the external magnetic field B. The red dashed horizontal line shows the absolute value of the magnetic susceptibility of gem-quality diamond $|\chi_{dm}| = 2 \cdot 10^{-5}$ SI units.

$$\chi_{fr}(\zeta, T) = \frac{N_{fr}(\zeta g_r \mu_B)^2 \mu_0}{4 k_B T} \frac{(2+\zeta)}{3\zeta}. \tag{13}$$

We note that for fixed concentration of single spin radicals $N_r = \zeta N_{fr}$ and large values of ζ, equation (13) demonstrates a linear increase in the magnetic susceptibility χ_{fr} with an increase in the number $\zeta \gg 1$ of uncompensated spin radicals per ferron ($N_{fr} = N_r/\zeta$ is the concentration of ferrons).

3. Application to the analysis of magnetic susceptibility behavior in diamond with radiation-induced defects

The magnetic susceptibility χ_{dm} of natural gem-quality diamonds (defect-free) is negative (i.e., they are diamagnetic) and practically temperature-independent, at least for $T < 300$ K [24]. Using the value $\chi_\rho = \chi_{dm}/\rho = -4\pi \cdot 0.446 \cdot 10^{-6}$ cm^3 g^{-1} from [24] and the diamond crystal density $\rho \approx 3.52$ g cm^{-3} [25], we obtain: $\chi_{dm} = \rho\chi_\rho = -2 \cdot 10^{-5}$ SI units. This experimental value χ_{dm} finds support in calculations from first principles [26].

Figures 2 and 3, respectively, illustrate the magnetic-field dependence (at $T = 3$ K) and temperature dependence (at $B \to 0$) of the differential magnetic susceptibility for the 'gas' of spin radicals $\chi_r \equiv \chi_{fr}(\zeta = 1)$ according to equation (5), as well as their clusters (ferrons) $\chi_{fr}(\zeta)$ according to equation (10) for the number of uncompensated spin radicals per a ferron $\zeta = 10, 30, 100, 200, 350$. A typical concentration of spin radicals $N_r = 10^{19}$ cm^{-3} is used, which is characteristic for experiments on radiation damage of diamond where its crystalline structure is still preserved.

As seen in figure 2 (blue curve), for ferrons with $\zeta = 200$ at temperature $T = 3$ K, their spin contribution to the susceptibility compensates the diamagnetic contribution from the diamond matrix χ_{dm} (red line) in fields below 0.4 T. Moreover, the larger the number ζ of spin radicals in each ferron, the lower the induction B at which the low-temperature magnetic susceptibility χ_{fr} due to the gas of ferrons is greater than the susceptibility χ_r of the gas of radicals ($\zeta = 1$; black dashed line).

Figure 3 shows that the mutual compensation of $\chi_{fr} > 0$ and $\chi_{dm} < 0$ for $\zeta = 200$ in the limit $B \to 0$ occurs at $T \approx 200$ K (the intersection point of the blue and red lines). This observation finds support in experiment [7], where the magnetic susceptibility of partially disordered neutron-irradiated natural diamonds becomes zero at $T \approx 150$ K.

There are experimental grounds [27–29] to believe that in a ferron the local density of unpaired (uncompensated) electron spins does not exceed a certain limiting value $N_{cr} = 10^{20}$ cm^{-3}. Then, in the approximation of a spherically shaped ferron containing ζ uncompensated spins, its diameter is

$$d_{fr} = \left(\frac{6\zeta}{\pi N_{cr}}\right)^{1/3}. \tag{14}$$

For a spin count per ferron $\zeta = 10$–350, equation (14) yields an average size of ferrons $d_{fr} = 6$–19 nm.

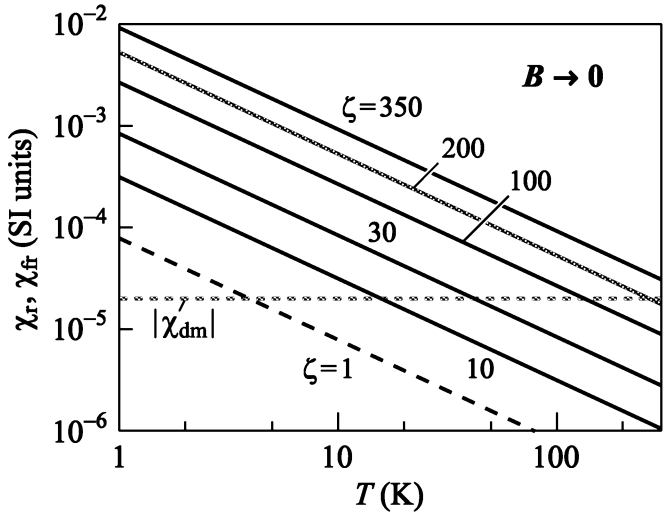

Figure 3. Temperature dependence of differential magnetic susceptibility of isolated spin radicals $\chi_r \equiv \chi_{fr}$ for $\zeta = 1$ by equation (5) and ferrons (spin-radical clusters) $\chi_{fr}(\zeta, T)$ by equation (13) at $B \to 0$ for $\zeta = 10, 30, 100, 200, 350$, and spin-radical concentration $N_r = 10^{19}$ cm^{-3} in partially disordered diamond. The red dashed horizontal line shows the absolute value of the magnetic susceptibility of gem-quality diamond $|\chi_{dm}| = 2 \cdot 10^{-5}$ SI units.

Calculations using the proposed model with reference to figures 2 and 3 for $T = 3$ K, $\zeta = 200$, and $d_{fr} \approx 16$ nm give an estimated susceptibility $\chi \approx 1.8 \cdot 10^{-3}$ SI units at $B \to 0$. This estimate is in good agreement with the value $\chi = 1.8 \cdot 10^{-3}$ SI units measured in [7] at $T \approx 3$ K for neutron-irradiated (fluence of $\approx 5 \cdot 10^{20}$ cm^{-2}) natural type IIa diamond crystals (samples comprises 113 rounded grains, each ≈ 0.5 mm in diameter). Magnetic measurements in the range of magnetic field B from -8 to 8 T were carried out on a High Field Cryogen Free Measurement System with a Vibrating Sample Magnetometer (Cryogenic Ltd., London, UK) in the temperature range from 2 to 300 K. Under experimental conditions, the concentration of both conduction band electrons and valence band holes in the studied samples was much lower than the concentration of spin radicals. Note that in the similar diamond grains not irradiated with neutrons (113 pieces), only diamagnetic susceptibility was observed over the entire range of temperatures and magnetic fields studied [7].

According to [24], the ferromagnetism in diamond arises from interspin exchange interactions, leading to the ordering of magnetic moments of impurity atoms (or intrinsic defects of the crystal structure). This phenomenon is characteristic of their associates (clusters) exceeding 10 nm in size.

It has been established (see [30] and references therein) that in diamonds, with increasing fluence of ionizing radiation, the Fermi level (E_F) shifts to a limiting position ≈ 1.8 eV above the valence band edge. This stabilization (or pinning) of E_F is attributed to the accumulation of radiation defects of the same type, each of which can exist in three charge states ($-1, 0, +1$; in units of elementary charge). Such radiation defects in the crystal structure are immobile at temperatures below 400 °C and are sufficient to ensure the electrical neutrality of partially disordered diamond. However, due to thermally activated electron hopping between these defects, their charge states migrate through the crystal and can form ferrons. In [28], it was found that in natural diamond samples irradiated with fast reactor neutrons (as in [7]), the thermal activation energy of DC hopping electrical conductivity is ≈ 170 meV (in the temperature range of 30–300 °C). Valence band holes and conduction band electrons are absent in partially disordered diamond (band gap ≈ 5.45 eV) even at room temperature, and thus cannot contribute to the magnetism of such diamond. In contrast, clustering-enhanced spin-radical magnetism for the number of spin radicals in a cluster (ferron) $\zeta > 200$ persists up to room temperature (see figure 3).

Finally, we note that the electron spins of radicals in ferrons can be partially compensated (antiferromagnetic ordering). This circumstance can be taken into account by introducing a fraction $\nu \leqslant 1$ of magnetoactive (uncompensated) spins in a ferron. Then, the concentration of non-interacting ferrons in the second sample remains the same $N_{fr} = N_r/\zeta$, where N_r is the concentration of spin radicals in the first sample. However, the total spin magnetic moment of ferron changes, becoming equal to $\nu\zeta g_r \mu_B/2$. To account for this issue, ζ should be replaced by $\nu\zeta$ in formulas (7)–(13). Notably, formula (14) for the ferron diameter remains unchanged. The number ζ in formula (14) represents the total number of radicals with uncompensated spins 1/2 (when radicals are in a superparamagnetic state) and with compensated spins (when radicals are in an antiferromagnetic state). In the calculations presented in figures 2 and 3, ν was set to 1, meaning all radical spins in the ferron are uncompensated [31].

4. Conclusions

The effect of magnetization enhancement in crystalline material has been theoretically studied, leading to manifestations of superparamagnetism due to the formation and growth of ferrons. These are clusters of uncompensated magnetic moments originating from structural defects (spin radicals). It was assumed that the concentration of localized spin radicals significantly exceeds the concentration of conduction-band electrons and valence-band holes. Consequently, the contributions to magnetic susceptibility of diamond samples from delocalized charge and spin carriers were disregarded in the analysis.

The magnetization and spin susceptibility of diamond were determined using the Brillouin function applied both to the case of no clustering (where the sample contains only identical isolated spin radicals exhibiting paramagnetic behavior) and to cases where the spin radicals form isolated spherical clusters—ferrons. Low-temperature magnetic susceptibility was calculated based on the Brillouin function. The study demonstrates that at helium temperatures in the limit of zero induction of the external magnetic field, the formation and growth of ferrons enhance superparamagnetic properties: magnetic susceptibility increases by two orders of magnitude compared to isolated spin-radical system.

The predictions of the theoretical model were compared with the experimental results on the low-temperature magnetic susceptibility of reactor-neutron-irradiated natural IIa diamonds. The parameters considered were ζ and N_r. The number of spin radicals per cluster ζ varied from 10 to 350 while maintaining the average spin-radical density $N_r = 10^{19}$ cm^{-3} in the samples. For the limiting value of the spin-radical density in a ferron, its experimental estimate was used: 10^{20} cm^{-3}. The results on the magnetic susceptibility calculation closely match the experimental data for crystalline grains of neutron-irradiated natural IIa diamond [7], allowing to estimate the number of uncompensated spin radicals per ferron as 200 and corresponding ferron size as 16 nm. These findings highlight the potential for developing methods to form ferrons by exposing initially non-magnetic diamonds to ionizing radiation for applications in spin-based cryoelectronics components.

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Conflict of interest

The authors declare no conflict of interest.

Data availability statement

The data that support the findings of this study are openly available at the following URL/DOI: https://doi.org/10.17632/9br5gzjtg4.

Author contributions

N A Poklonski 0000-0002-0799-6950
Conceptualization (equal), Funding acquisition (lead), Investigation (equal), Methodology (lead), Writing – original draft (lead)

S A Vyrko 0000-0002-1145-1099
Investigation (equal), Software (lead), Visualization (lead), Writing – review & editing (equal)

A G Zabrodskii 0000-0002-4073-3079
Conceptualization (equal), Writing – review & editing (equal)

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