# Investigation of the superparamagnetic properties of ultrafine iron particles by Mössbauer spectroscopy

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The superparamagnetic properties of 105 to 230Å diameter iron particles are investigated by using a dilute system at temperatures of 80 and 300°K and with a nondilute system at temperatures ranging from 300 to 1100°K in a vacuum furnace. The effect of a 3-kOe external magnetic field on the dilute system is studied. A modified semi-empirical paramagnetic formula is employed, which permits one to calculate the pre-exponential factor for arbitrary particle size and which takes into account interaction between the particles and between the particles and the external magnetic field. It is found that the effective anisotropy decreases for iron particles with sizes exceeding a certain critical value corresponding to deviation from parallel distribution of the magnetic moments in the particles. A comparison of the experimental data and theory yields $d_{cr} = 95 \pm 25$Å. The effective Curie point for the particles decreases and their magnetic dipole interaction energy at 700–850°K also is found to decrease. The effective constant for mutual interaction between the particles and in an external magnetic field is determined and found to be $\sim 0.1$. The strong reduction of magnetic interaction between the particles is ascribed to the presence of an oxide on the particle surface.

Ultrasmall particles of magnetically-ordered substances exhibit a number of unique magnetic properties. These include the presence in the particles of thermal fluctuations that lead to the vanishing of the average magnetic moment in the sample. This phenomenon was named superparamagnetism in 1956[1]. The frequency of the fluctuation of the magnetic moment[2] is determined by the formula

$$f = f_0 \exp (-Kv / kT), \qquad (1)$$

where $f_0 \sim 10^{10}$ Hz, $K$ is the anisotropy constant of the substance along the easy-magnetization axis, $v$ is the volume of the particle, and $T$ is the temperature. The quantity $f$ depends both on the magnetic properties of the substance (via the constant $K$) and on the particle dimension. As a result, the fluctuation frequency and the magnetic-moment relaxation time $\tau$ can vary in a wide range from 1 to $10^{-10}$ sec. Whether a magnetically-ordered substance exhibits superparamagnetic properties depends essentially on the measurement time. A convenient method for the study of the features of magnetic-moment relaxation in ultrasmall particles with dimensions $\sim 100$ Å, for which $\tau = 10^{-7}$–$10^{-10}$ sec, is therefore $\gamma$-resonance (GR) spectroscopy, with the aid of which it is possible to investigate the transition to superparamagnetism as a function of the volume of the particles and the temperature, within times that are comparable with the period of the Larmor precession of the employed nucleus. For example, for $Fe^{57}$ in a field $H = 500$ kOe, the nuclear-spin Larmor precession time is $10^{-8}$ sec.

Besides its use for the study of magnetization, GR spectroscopy has recently become the principal method of studying superparamagnetism[3-22]. The research was carried out mainly along the following directions: 1) study of the effective lowering of the Curie or Neel point in ultrasmall particles in comparison with bulky material; 2) determination of the anisotropy constants in small particles; 3) calculation of the particle distribution by sizes; 4) investigation of the magnetic properties of the atoms on the surface and in the volume of the particles and of the dependences of the magnetic properties of the particles on the dimensions.

It was previously predicted theoretically[19,20] that a new effect exists in the superparamagnetic region and consists of a sharp decrease in the relaxation time for particles of a substance having a cubic crystal lattice. Starting with a certain particle dimension $d = d_{cr}$, the magnetization of the particles becomes inhomogeneous because the movements of the peripheral atoms cease to be parallel, and the effective anisotropy constant reverses sign. An experiment was performed[19,20] on particles of an FeNi alloy (37% Ni), and has shown qualitative agreement with the theory. In this case, however, only two dimensions could be obtained in the required range of particle dimensions, so that the predicted effects could not be investigated.

The present investigation of superparamagnetism was undertaken for the purpose of studying the features of superparamagnetism with ultrasmall particles of iron having different dimensions, inasmuch as bulk iron is the most thoroughly studied magnetic material. We investigated the influence of the external magnetic field on the magnetic-relaxation properties of the particles and took into account the magnetic interaction of the particles with one another.

## EXPERIMENTAL PROCEDURE AND RESULTS

The iron particles were produced by an aerosol method and a procedure described in[19,20]. The particle dimensions were determined with an electron microscope. After plotting the particle distribution with respect to the dimensions, we determined the mean cubic diameter of the particle. As a result we obtained four initial particle dimensions: $125 \pm 20$, $170 \pm 25$, $195 \pm 35$, and $260 \pm 40$ Å. The particles were coated with a layer of $\gamma$-oxide of iron[21]. The number of atoms of oxidized iron and the number of layers of oxide were estimated from the corresponding areas under the GR spectra of the iron and of the oxide at $T = 80°$K[21]. In addition, particles with $d = 125$ Å were investigated in

two variants: oxidized in air for several months, and freshly oxidized in several days. In the latter case, the iron oxide layer on the particle surface was thinner. Thus, the dimension of the particles of the metallic iron proper were $d_1 = 105 \pm 25$ Å, $d_2 = 115 \pm 25$ Å, $d_3 = 140 \pm 30$ Å, $d_4 = 165 \pm 40$ Å, $d_5 = 230 \pm 45$ Å. The particles with dimensions $d_1$ and $d_2$ were obtained from particles having an initial dimension 125 Å. The GR spectra were obtained in the temperature range from 80 to 1100° K. Above 300° K, the particles were investigated in a special vacuum oven, in which the vacuum at 1100° K was not worse than $10^{-3}$ mm Hg. During the time of the experiment, the particles were located between two beryllium disks. Inasmuch as the beryllium contained a small admixture of iron, the line corresponding to the impurity was subtracted as a background from the summary spectrum. The experiments below 20° K were performed in a cryostat, in which a magnetic field of 3 kOe perpendicular to the direction of the $\gamma$ quanta could be produced. For low-temperature experiments, we used strongly diluted systems, $\sim 1\%$ by weight, consisting of iron particles and a filler. A more convenient system, which made it possible to separate the particles from one another in part, was a saturated solution of rosin in alcohol. Owing to the low boiling temperature of this system ($\sim 100°$ C), the iron particles were not oxidized (the boiling point was 5—10 min), and at the same time the boiling contributed to the separation of the particles from one another. All the measurements were performed with a GR spectrometer with constant acceleration and with a source of $Co^{57}$ in Cr. The isomer shifts were reckoned from the metallic iron.

Figure 1 shows the GR spectra of particles making up a dilute system at T = 300° K. The spectra consist of the hyperfine structure (hfs) of the metallic iron and of the iron-oxide impurity. The hfs lines of $\alpha$-Fe broaden with decreasing particle dimension, with the exception of the particle with d = 105 Å, the spectrum of which recalls the spectrum of the particles with d = 140 Å. Figure 2 shows the GR spectra of a dilute system of particles at temperatures 300 and 80° K in an external magnetic field H = 3 kOe. Lowering the temperature and application of

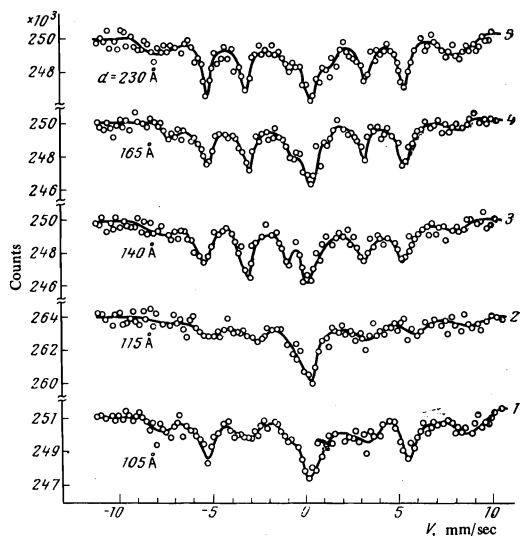the external magnetic field H = 3 kOe lead to stabilization of the hfs and to a narrowing of the $\alpha$-Fe lines, thus offering evidence of superparamagnetic behavior of the particles. Figure 3 shows the spectra of the particles in the vacuum oven in the temperature region 850° K, which indicates lower relaxation times in comparison with the spectra of Fig. 1. With decreasing particle dimension, the hfs vanishes, but for the particles with d = 105 Å, just as in the case of the dilute system, the $\alpha$-Fe line width is similar to the width of the hfs lines for the particles with d = 140 Å. As a result, the data of Figs. 1—3 prove that the iron particles have a superparamagnetic behavior that depends on their dimensions, whereas for particles with d = 105 Å it is observed that the relaxation time of the magnetic moment of $\alpha$-Fe increases and the hfs lines decrease.
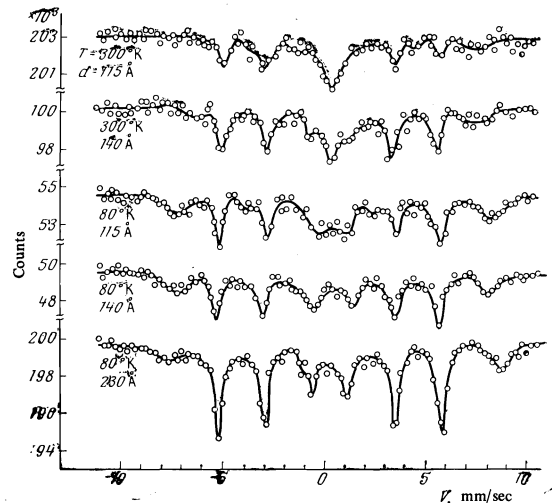


FIG. 2. GR spectra of ultrasmall iron particles at T = 80° K and T = 300° K in an external magnetic field H = 3 kOe.
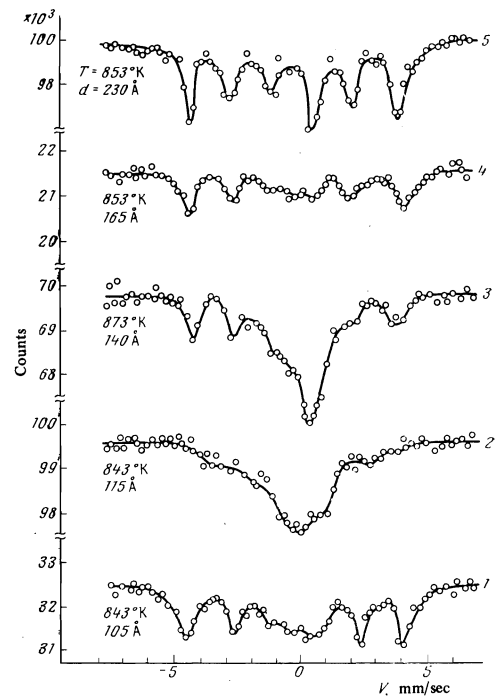


FIG. 3. GR spectra of ultrasmall iron particles with diameters d = 105 − 230 Å at T $\sim$ 850° K. The particle relaxation time is r: $1 - \tau = (1 \pm 1) \cdot 10^{-8}$ sec, $2 - \tau = (2 \pm 1) \cdot 10^{-9}$ sec, $3 - \tau = (6 \pm 2) \cdot 10^{-9}$ sec, $4 - \tau = (1 \pm 1) \cdot 10^{-8}$ sec, $5 - \tau = 1.4 \cdot 10^{-7}$ sec.



FIG. 1. GR spectra of ultrasmall iron particles with diameters from 105 to 230 Å at T = 300° K. The particle magnetic-moment relaxation times are: $1 - \tau = (2.8 \pm 1.1) \cdot 10^{-8}$ sec, $2 - \tau = (9.8 \pm 1.1) \cdot 10^{-9}$ sec, $3 - \tau = (1.6 \pm 0.4) \cdot 10^{-8}$ sec. $4 - \tau = (2.8 \pm 1.1) \cdot 10^{-8}$ sec, $5 - \tau = (14.4 \pm 2.9 \cdot 10^{-8}$ sec.

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## DISCUSSION OF EXPERIMENTAL RESULTS

Our experiments make it possible to analyze the superparamagnetic properties of particles in the absence of a magnetic dipole interaction between the particles, and to investigate the influence of the external magnetic field and the temperature as well as the influence of the interaction between the particles.

### 1. Modified semiempirical superparamagnetism formula

Since the principal parameter determined in our experiments is the relaxation time $\tau$ of the magnetic moment of the particle it is necessary to have a formula connecting $\tau$ with the parameters of the superparamagnetic particle. In the absence of interaction between the particles we have

$$\tau = \tau_0 \exp(Kv / kT), \qquad (2)$$

where

$$\tau_0 = (M_s / \gamma_0 K) \sqrt{kT / Kv} \quad \text{at} \quad Kv / kT \geqslant 2$$

(here $M_S$ is the saturation magnetic moment and $\gamma_0$ is the gyromagnetic ratio[23]).

This formula, however, becomes meaningless when $K$ or $v = 0$. On the other hand, it does not take into account the action of the external magnetic fields and the interaction of the particles with one another. We can modify formula (2) somewhat, with allowance for the shortcomings listed above, by considering the following model. At $Kv = 0$, we shall regard the particle as paramagnetic and in which the relaxation time is determined by the spin-spin relaxation. It is easy to show in this case that

$$\tau = \tau_0 = ha^3 / n^2 \mu_B^2, \qquad (3)$$

where $a$ is the distance between the paramagnetic atoms, $n$ is the number of Bohr magnetons per atom, and $\mu_B$ is the Bohr magneton. This definition of $\tau_0$ no longer depends on $K$ or on $v$. When the finite quantity $Kv$ appears (when magnetic ordering of the volume $v$ sets in), an exponential term appears in (3) and leads to a slowing down of the relaxation, in analogy with formula (2). In the case of interaction between the particles, the potential barrier $U = Kv$, which hinders the relaxation of the magnetic moment, acquires a term $\alpha MH_{\text{eff}}$, which takes into account their magnetic dipole interactions with one another, where $M$ is the magnetic moment of one particle, $H_{\text{eff}}$ is the effective magnetic field of all the remaining particles, and $\alpha$ is the particle interaction constant. Assuming $H_{\text{eff}} \approx M/d^3$, where $d$ is the particle diameter, we can express the energy of the interaction of the particles with one another in the form $\alpha M^2/d^3$. In the presence of an external magnetic field, the potential barrier that hinders the rotation of the magnetic moment acquires an additional term $\beta MH$. The resultant final formula is

$$\tau = \tau_0 \exp\left(\frac{U}{kT} + \frac{\alpha M^2/d^3}{kT} + \frac{\beta MH}{kT}\right), \qquad (4)$$

$$\tau_0 = \frac{ha^3}{n^2 \mu_B^2},$$

Thus, this formula makes it possible to estimate the quantities $U$, $\alpha$, $\beta$, and $M$, which characterize the magnetic properties of the particle, from the GR spectra in the range $\tau \sim 10^{-7}-10^{-10}$ sec.

### 2. Influence of decreasing the effective anisotropy near $d = d_{cr}$

The anisotropy energy was determined both from experiments with a dilute system of particles and with an undiluted system at high temperatures. For the dilute system, it was assumed that the magnetic dipole interaction between the particle is small in comparison with $kT$ and that $\alpha M^2/d^3 \approx 0$ in formula (4). For the undiluted system at high temperature, as will be shown below, the particle interaction energy becomes of the order of $kT$, so that the term $U$ in (4) comes into play. Since $U$ is comparable with $kT$ at high energies, it was assumed in the calculation of $U$ from the high-temperature experiment that $\alpha M^2/d^3 \approx U$.

The relaxation time was determined from the relaxation broadening of the line[24]:

$$\tau = \hbar / \Gamma, \qquad (5)$$

at $\Delta\Gamma \ll H_{\text{nuc}}$ ($H_{\text{nuc}}$ is the magnetic field at the nucleus), with the non-relaxation values of $\Gamma$ assumed to be the experimental half-width of the hfs line of the particles with dimensions 230 Å at 80° K in an external magnetic field of 3 kOe (Fig. 1) and that obtained from the shape of the spectrum as a function of $\tau$ at $H_{\text{nuc}}$ = 330 kOe.[25] In the calculation of $\tau$, the value of $\tau_0$, which we determined from (4), was $3 \times 10^{-10}$ sec. The experimental results of the calculation of $U$ from formula (4) for the diluted system at $T = 300°$ K and for the undiluted system at $T = 850°$ K are shown in Fig. 4.

We turn now to a comparison of the experimental data with the qualitative theory developed in[19,20]. With increasing diameter of the homogeneously magnetized particle, the quantity $U$ increases rapidly; this is shown dashed in Fig. 4 at $K = 6 \times 10^5$ erg/cm$^3$. However, starting with a set dimension $d = d_{cr}$, an anomaly appears in the behavior of $U$. The effective isotropy $U$ decreases and reverses sign, so that a V-shape dependence of $U$ on the particle dimension is obtained. This effect is connected with the start of the deviation from the homogeneous magnetization for a single-domain particle, as a result of the rotation of the magnetic moments of individual atoms at the start of the domain formation.

The dependence of $U$ on the particle diameter was calculated in accordance with the formula (see[19,20])

$$U = \tfrac{1}{2}K \int (\cos^4\omega - 3\cos^2\omega\sin^2\omega + \tfrac{3}{8}\sin^4\omega)\,dv. \qquad (6)$$

Here $\omega(r, \theta) = \text{cf}(z)\sin\theta$, $f(z) = z^{-1/2}J_{3/2}(\lambda_0 z)$, $z = r/R$; $\lambda_0 = 2.085$.

At $z = 1$ we have

$$d_{cr} = 2a\sqrt{A} / I, \qquad (7)$$

where $a = 1.04-1.44$ is the demagnetization factor and depends on the shape of the particle, $A$ is the value of the exchange integral, and $I$ is the saturation magnetization of the substance. At $\omega = 0$, the particle is homogeneously magnetized. We note that calculation in accordance with formula (6) is valid only near $d_{cr}$, in the region $d \lesssim 2d_{cr}$[19,20], and is qualitative in character. Figure 4 shows the calculated dependence of $U$ on the particle diameter, obtained for $d_{cr} = 95 \pm 25$ Å. There are apparently several reasons for the quantitative deviation of the experimental results from the theory: these include the dispersion of the particle sizes, devia-
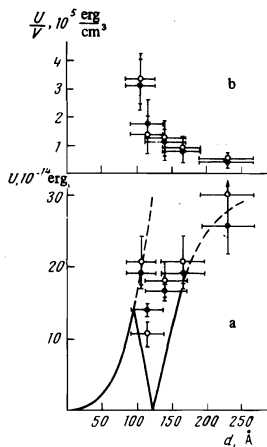
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FIG. 4. a) Experimental and calculated values of U as functions of the dimension of metallic-iron particles at T = 300°K (dark circles) and T ≈ 850°K (light circles). The dashed lines show the function U = Kv/2 without allowance for the theory of [19,20].
b) Experimental values of K/2 = U/v as functions of the metallic-iron particle dimensions. The symbols are the same.



FIG. 5. Temperature dependence of the effective magnetic field $H_{nuc}$ at the $Fe^{57}$ nucleus for different particle dimensions (1—105Å, 2—115Å, 3—140Å, 4—105Å, 5—230Å).

tions from spherical shape, and the deviation of the value of $\tau_0$ from that assumed in this paper. Nevertheless, even this experiment makes it possible to estimate the exchange integral for iron: $A = (0.35^{-0.28}_{-0.12})$ × $10^{-6}$ erg/cm.

In the calculation of A we assumed I = 1700 G and a = 1.44 for a sphere. However, taking into account the possibility of as much of a 20% deviation of the particle shape from spherical, the value of a can decrease to $1.3^{[26]}$, a fact accounted for in the determination of the error of A. The value of A agrees with data determined by other methods[27]. Thus, the decrease of the effective anisotropy can serve as a new independent method of determining A. We call attention to the fact that the effective anisotropy in the range 300—850°K remains constant within the limits of errors. At the same time, data on the dependence of the anisotropy constant $K = f(T)^{[26]}$ indicate that K decreases by at least one order of magnitude at 850°K in comparison with 300°K. It should be noted that the values of K are usually determined with the aid of single crystals with an anisometer, and characterize the macroscopic properties of the substance. Inasmuch as the substance begins to break up near $T_C$ into a series of superparamagnetic regions that interact weakly with one another, this can lead to an overall decrease of K. The local microscopic effective anisotropy U, however, remains apparently unchanged, as is evidenced by our data.

Figure 4 shows also the experimental values of K/2 = U/v. The value of K decreases with increasing dimension and with deviation of the state of the particle from homogeneous magnetization, in qualitative agreement with data on the measurement of the coercive force[26].

## 3. Influence of particle magnetic-dipole interaction and of external magnetic field on the magnetic-moment relaxation time in the particles

Figure 5 shows the values of the effective magnetic field $H_{nuc}$ at the iron nuclei as functions of the temperature for different particles. For an undiluted system, in the range up to 700° K, no relaxation phenomena are observed in the spectra, and no superparamagnetism appears. This is due to the magnetic dipole interaction of the particles with one another, as a result of which the relaxation is hindered and $\tau$ in formula (4) increases on account of the term $\alpha M^2/d^3$. The temperature dependence of $H_{nuc}$ is therefore the same for all particles up to 700° K. With increasing temperature, differences appear,
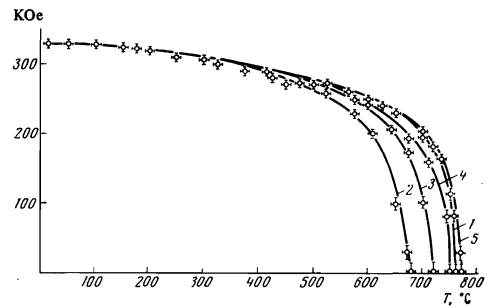
for example, for the 115-Å particles the deviations of the $H_{nuc} = f(T)$ dependence from the data for bulky material are observed at 700° K, and for the 165-Å particles they appear at 850° K. The effective Curie points $T_{C.eff}$ determined by us for the particles are 1035 ± 5°, 1025 ± 5°, 1020 ± 5°, 1000 ± 5°, and 945 ± 5° K for particles with d = 230, 105, 965, 140, and 115 Å, respectively. If we trace the variation of $T_{C.eff}$, we see that we have again an anomalous increase of $T_{C.eff}$ for 105-Å particles in comparison with 115-Å particles, in analogy with the dependence of U on the particle diameter at T = 300 and 900° K. This circumstance confirms once more the fact that the magnetic dipole interactions become of the order of kT or smaller and the spin relaxation is determined by the dependence on the particle diameter.

The temperature at which the $H_{nuc} = f(T)$ dependence for any particular particle size begins to deviate from the dependence for the bulky material corresponds to the energy at which the magnetic interaction energy between the particles becomes of the order of kT. We make use of this fact to estimate the effective magnetic moment of the particle and the interaction constant $\alpha$ from $\alpha M^2/d^3 \approx kT$. For particles with d = 115 Å we have $\sqrt{\alpha}M = 4.5 \times 10^{-16}$ erg/G, and for particles with d = 165 Å we get $\sqrt{\alpha}M = 9.8 \times 10^{-16}$ erg/G, which yields $\alpha \approx 0.2$ and $\alpha \approx 0.1$, respectively. In the calculations we assumed an iron-lattice constant a = 2.86 Å and n = 2.2 Bohr magnetons per atom.

The influence of an external magnetic field H = 3 kOe was investigated with a dilute system at $\alpha M^2/d^3 \approx 0$; as follows from Fig. 2, a narrowing of the line and an increase of the relaxation time take place at 300° K, and a complete stabilization of the hfs occurs at T = 80° K (the small dip in the center of the spectrum is probably due to the spectrum of the iron oxide on the particle surface). Using the relaxation broadenings of the outermost hfs spectral lines of iron with d = 115 Å at H = 3 kOe (Fig. 2) as well as the previously determined value U at H = 0, we estimate the value of $\beta MH$. The result is $\beta M = 0.5$ × $10^{-16}$ erg/G, so that $\beta \sim 0.1$. Thus, both the interaction between the particles, and their interaction with the external magnetic field are weakened by one order of magnitude in the case of iron particles with dimension ~100 Å. This result agrees with the data of[28], where the magnetic moment of Fe, Co, and Ni particles was investigated by investigating the particle magnetization.

It was found that for ~80-Å particles of Fe and Co the magnetic moment amounts to 20% of the moment of the bulky material, this being attributed to the influence of the oxide on the particle surface. It is also known that surface exchange anisotropy with the atoms of the metal

exists for CoO and NiO on the surface of Co and Ni particles[26] and leads to an increase of the coercive force. In our case, the presence of oxide on the surface hinders the orientation of the magnetic moment in the external magnetic field and hinders parallel orientation of the magnetic moments of the particles relative to one another. The same causes can hinder the thermal fluctuations of the magnetic moment in small particles, as a result of which U increases in comparison with the theory (Fig. 4).

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[1] C. P. Bean, J. Appl. Phys. **26**, 1381, 1955.

[2] L. Neel, C. R., Paris, **228**, 664, 1949.

[3] T. Nakamura T. Shinjo, Y. Endon, N. Yamamoto, M. Shiba, and Y. Nakamura, Phys. Lett. **12**, 178, 1964.

[4] T. Shinjo, J. Phys. Soc. Jap. **21**, 917, 1966.

[5] S. Nazu, T. Shinjo, Y. Nakamura, and Y. Murakani, J. Phys. Soc. Jap. **23**, 1001, 1968.

[6] K. I. Ando, W. Kündig, G. Constabaris and R. M. Lindquist, J. Phys. Chem. Sol. **28**, 2291, 1967.

[7] A. M. van der Kraan, and I. van Loef, Phys. Lett. **20**, 614, 1966.

[8] W. Kündig, H. Bömmel, G. Constabaris and R. Lindquist, Phys. Rev. **142**, 327, 1966.

[9] W. I. Schuele, S. Shtrikman and D. Treves, J. Appl. Phys. **36**, 1010, 1965.

[10] U. Gonser, H. Wiedersich and R. W. Grant, J. Appl. Phys. **39**, 1004, 1968.

[11] R. N. Lindquist, G. Constabaris, W. Kündig and A. M. Portis, **39**, 1001, 1968.

[12] M. Eibschütz and S. Shtrickman, J. Appl. Phys. **39**, 997, 1968.

[13] M. Rone, H. Schechter and S. Needzweedz, J. Appl. Phys. **39**, 265, 1968.

[14] W. Kündig, M. Kobelt, H. Appel, G. Constabaris and R. H. Lindquist, J. Phys. Chem. Sol. **30**, 819, 1969.

[15] J. M. D. Coey, M. D. Spender and A. H. Morrish, Sol. State Com. **8**, 1605, 1970.

[16] S. M. Aharony and M. H. Litt, J. Appl. Phys. **42**, 352, 1971.

[17] J. M. D. Coey and D. Khalafalla, Phys. St. Sol., a, **11**, 229, 1972.

[18] I. P. Suzdalev, Fiz. Tverd. Tela **12**, 988 (1970) [Sov. Phys.-Solid State **12**, 775 (1970)].

[19] A. M. Afanas'ev, I. P. Suzdalev, M. Ya. Gen, V. I. Gol'danskiĭ, V. P. Korneev, and E. A. Manykin, Zh. Eksp. Teor. Fiz. **58**, 115 (1970) [Sov. Phys.-JETP **31**, 65 (1970)].

[20] A. M. Afanasiev, I. P. Suzdalev and E. A. Manikin, Proc. of the Conference on the Appl. of Mössbauer Effect (Tihany, Hungary, 1969), p. 183.

[21] I. P. Suzdalev and A. P. Amulyavichus, Zh. Eksp. Teor. Fiz. **63**, 1758 (1972) [Sov. Phys.-JETP **36**, 929 (1973)].

[22] A. M. van der Kraan, Thesis, Krips-Repro N. V.—Meppel, Holland, 1972.

[23] A. Aharony, Phys. Rev. **135A**, 447, 1964.

[24] A. M. Afanas'ev and Yu. Kagan, Zh. Eksp. Teor. Fiz. **45**, 1660 (1963) [Sov. Phys.-JETP **18**, 1139 (1964)].

[25] H. H. Wickman, M. P. Klein and D. A. Shirley, Phys. Rev. **152**, 345, 1966.

[26] S. V. Vonsovskiĭ, Magnetizm (Magnetism) Nauka, 1971, pp. 416, 785, 787, 790.

[27] E. H. Frei, S. Shtrikman and D. Treves, Phys. Rev. **106**, 447, 1957.

[28] A. Tasaki, S. Tomiyama, S. Iida, N. Wada, and R. Uyeda, Jap. J. Appl. Phys. **4**, 707, 1965.