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Theory for the atomic shell structure of the cluster magnetic moment and magnetoresistance of a cluster ensemble

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Abstract. We present a simple theory for the cluster size dependence of the average cluster magnetic moment of transition metal clusters. Assuming a local environmental dependence of the atomic magnetic moments, the cluster magnetization exhibits a magnetic shell structure, reflecting the atomic structure of the cluster. Thus, the observed oscillations of the average cluster magnet moment may serve as a fingerprint of the cluster geometry. We also discuss the giant magnetoresistance (GMR) exhibited by an ensemble of magnetic clusters embedded in a metallic matrix. It is shown that the magnetic anisotropy affects strongly the magnetization of the cluster ensemble under certain conditions. Since the GMR depends on the cluster ensemble magnetization, it can be used to determine the cluster magnetic anisotropy energy.

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1 Introduction

Magnetic clusters, in particular transition metal and rare earth metal clusters, have been studied intensively both experimentally [1–4] and theoretically [5], and exhibit interesting magnetic properties. Recent Stern– Gerlach– experiments revealed oscillations in the magnetization of small Fe, Co, and Ni clusters as a function of the cluster size in the size range $50 < N < 700$ [1, 2]. The local magnetic moment, in particular for itinerant electron systems, depends sensitively on its atomic environment. As a rule of thumb the magnetic moment is the larger the lower the coordination number [6]. Therefore, the average cluster magnetic moment per atom of transition metal clusters is expected to vary with cluster size and structure, and in particular to reflect the atomic cluster geometry. The interplay of the geometric and the magnetic structure has been calculated only for relatively small clusters [5]. In Sect. 2 we present a simple model for the calculation of the average cluster magnetic moment as a function of the cluster size and cluster atomic structure. The atomic magnetic moments are assumed to depend only on their local environment. Shell-by-shell cluster growth and different cluster structures are considered. The results calculated by this theory are compared with experimental data.

Furthermore, we analyze the magnetoresistance of an ensemble of magnetic metal clusters embedded in a metallic matrix. The magnetic moments of the cluster atoms are ferromagnetically aligned, if the cluster temperature is well below the internal magnetic ordering temperature of the cluster. Hence, when deposited on a metallic substrate or dissolved into a nonmagnetic metallic matrix, the magnetic clusters act like paramagnetic impurities with large magnetic moments and thus cause a giant magnetoresistance (GMR), as observed experimentally [7]. In Sect. 3 we discuss the possible connection between the magnetization, the magnetic anisotropy, and GMR of a cluster ensemble. The temperature and magnetic field dependence of GMR may be used to determine the magnetic anisotropy.

2 Average cluster magnetic moment

In this section we present a simple analysis of the atomic structure effects on the average cluster magnetic moment as a function of the cluster size. In accordance with observations at surfaces of bulk material, of thin films, and of magnetic alloys we assume that the magnetic moments μ_i depend solely on the atomic environment of the lattice site i. In particular, for transition metals μ_i is expected to be the larger the lower the number of nearest neighbors q_i [6]. For a first estimate, we simply assume a surface magnetic moment μ_{surf} for the atoms at the cluster surface, and the bulk value μ_{bulk} for the other atoms. Then the average cluster magnetic moment per atom is given by

$$\bar{\mu}(N) = (\mu_{\text{surf}} - \mu_{\text{bulk}})N^{-1/3} + \mu_{\text{bulk}} \,, \tag{1}$$

N being the number of cluster atoms. Such a behavior of $\bar{\mu}(N)$ should reflect the magnetic moments of very large clusters. For smaller clusters, especially in the size range of several hundred atoms [1, 2], details of the atomic cluster structure should be visible in the magnetic behavior. Then $\bar{\mu}(N)$ and the distribution of μ_i will reflect also the cluster geometry and lattice structure. From a comparison of $\bar{\mu}(N)$ with experimental data one may gain information about the cluster structure.

Table 1. Number of atoms N for various cluster structures with closed shells ('oct.'= octahedron, 'c.-o.'=cubo-octahedron). $N(\bar{\mu}_{\min})$ refers to the experimentally observed cluster sizes where the average magnetic moment $\bar{\mu}(N)$ is minimal [1]. Values in brackets refer to very shallow minima. n refers to the shell number, which is the length of the cluster edges in elementary units. For cubes and octahedrons the primed numbers n' refer to 'rounded' clusters, obtained by removing all edge atoms from clusters with n completely filled shells, yielding intermediate values of N. Comparison of the calculated values N with $N(\bar{\mu}_{\min})$ indicates that best agreement is obtained for cube shaped clusters with a b.c.c. lattice in the case of Fe, for cube shaped clusters with a f.c.c. lattice in the case of Ni, and for octahedral shaped clusters with a f.c.c. lattice in the case of Co

Shell	Closed shell cluster size N					$N(\bar{\mu}_{\min})$		
n	fcc-cube	fcc-oct.	fcc-c-o.	bcc-cube	bcc-oct.	Fe	Co	Ni
2'	43	43		15	27		50	
2	63	85	55	35	57	45	85	72
							129	
3'	140	165		59	89		173	131
3	172	231	147	91	143	85	232	175
4'	321	399		145	203	(150)	355	(260)
4	365	489	309	189	289	191	483	381
5'	610	777		285	385	273		625
5	666	891	561	341	511			
6'	1031	1331		491	651			
6	1099	1469	923	559	825	551		

Similarly as Billas et al. [2] we present first an atomic shell model for the average cluster magnetic moment. The cluster is assumed to grow shell-by-shell, occupying subsequently sites of a b.c.c. or a f.c.c. lattice. For larger clusters one expects that the cluster structure converges towards the bulk lattice structure, at least for the cluster interior. The overall cluster shape is expected to resemble a cube, an octahedron, or a cubo-octahedron, i.e. regular shapes which minimize the surface energy. In addition to such cluster shapes, we have considered a cluster growth assuming a succesive occupation of free sites with smallest distance from the cluster center, yielding compact and spherically shaped clusters. The magnetic moments are different for different atomic shells, and may even vary within the topmost incomplete shell. The average coordination number $\bar{q}$ will be largest for almost closed atomic shells. In accordance with the above mentioned general rule, maxima of $\bar{q}$ should correspond to minima of the average cluster magnetic moment $\bar{\mu}(N)$. Thus, an oscillatory behavior of $\bar{\mu}(N)$ as a function of the cluster size is expected by adding additional atomic shells to the cluster. This will modulate the monotonous behavior of $\bar{\mu}(N)$ as given by (1). Clearly, the oscillations of $\bar{\mu}(N)$ should reflect the atomic structure of the clusters.

To study also clusters with rounded edges, we assume for cubes and octahedrons also the following growth mode: first sites on the faces of the cluster surface are occupied by atoms, then after occupation of all face sites the edge sites are populated. The shell dependent magnetic moments are determined correspondingly.

Using these growth modes, we calculate the number of atoms N corresponding to various cluster sizes with closed atomic shells. Since for these sizes the average magnetic moment $\bar{\mu}(N)$ is expected to be minimal, we compare these numbers N with the measured $N(\bar{\mu}_{\min})$ for Fe, Co, and Ni clusters in the size range $30 < N < 700$ [1, 2], at which $\bar{\mu}(N)$ is minimal. Results are given in Table 1. For Fe the measured minima of $\bar{\mu}(N)$ seem to correspond best to N– values of closed shell sizes of a b.c.c.– cube. Similarly, the $\bar{\mu}$– minima of Ni clusters correspond best to N– values of closed shell f.c.c.– cubes, and the ones of Co clusters to f.c.c.– octahedrons. We have also considered the closed shell sizes of a number of other growth modes and atomic structures, in particular the f.c.c.– cubo-octahedron. The agreement with experimental data is not as good as for the above mentioned structures. Note that the results given in Table 1 allow certainly no definite conclusion about the cluster structure, but serve as an interesting hint and indicate which closed shell structure yield minima of $\bar{\mu}(N)$.

Next we calculate the average cluster magnetic moment $\bar{\mu}(N)$ by taking the atomic environmental dependence of the atomic magnetic moments more properly into account than in (1). First, a statistical model is proposed for the shell dependent magnetic moment and for the above mentioned growth modes. The average magnetic moment per atom of the topmost shell (0) is assumed to be

$$\bar{\mu}_0 = x_0(1 - x_0)\mu_{\text{at}} + x_0^2\,\mu_{\text{surf}}\,, \tag{2}$$

where x_0 is the concentration of statistically occupied sites in this shell. In (2), μ_{at} is the magnetic moment of an atom without nearest neighbors in the topmost shell, and μ_{surf} the magnetic moment of an atom surrounded by other atoms in the topmost shell, taken to be similar to the surface magnetic moment of bulk material. In addition, dependent on the concentration of deposited atoms x_0 in the topmost shell, the average magnetic moment $\bar{\mu}_1$ of the shell (1) below the topmost shell will also change. For the average magnetic moment of this shell we put

$$\bar{\mu}_1 = (1 - x_0)\mu_{\text{surf}} + x_0\,\mu_{\text{bulk}}\,. \tag{3}$$

Due to (3) the magnetic moment of an atom in shell (1) is equal to μ_{surf}, if it has no nearest neighbor atoms in the topmost shell (0), and is equal to the bulk value μ_{bulk}, if it is completely covered by topmost shell atoms. For simplicity, all magnetic moments of the inner atomic shells of the cluster are put equal to μ_{bulk}, simulating the increasing degree of itineracy and bulk character of the cluster interior. It follows from (2,3) that the average cluster magnetic moment per atom as a function of the cluster size N is given by

$$\bar{\mu}(N) = \frac{N_0\bar{\mu}_0 + N_1\bar{\mu}_1 + N_{\text{bulk}}\mu_{\text{bulk}}}{x_0N_0 + N_1 + N_{\text{bulk}}}\,, \tag{4}$$

with N_0 and N_1 being the number of sites in the two outermost cluster shells (0) and (1), dependent on the cluster shape, s. Table 1. N_{bulk} is the total number of atoms in the inner shells, and $N = x_0N_0 + N_1 + N_{\text{bulk}}$. Note that $\bar{\mu}(N)$ calculated using (4) yields a magnetic shell structure: it exhibits an oscillatory behavior with minima near closed cluster shells ($x_0 = 1$), and maxima for almost half filled shells, s. solid curves in Fig. 1. We assume in general $\mu_{\text{bulk}} < \mu_{\text{surf}} < \mu_{\text{at}}$.

Secondly, we assume that each magnetic moment μ_i on a lattice site i is determined by its actual number of nearest neighbors q_i. In particular, we put

$$\mu_i = \mu(q_i) = \begin{cases} \mu_1, & q_i \le q_c\,, \\ \mu_2, & q_i > q_c\,. \end{cases} \tag{5}$$

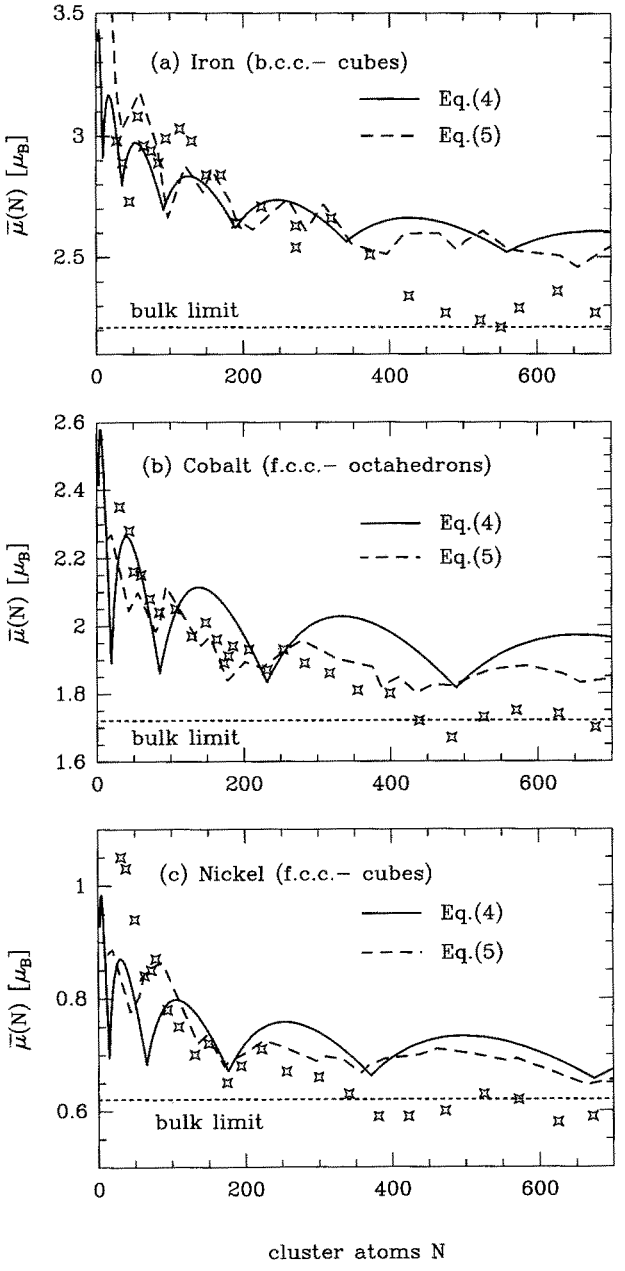

Fig. 1a–c. Results for the magnetic shell structure of the average magnetic moment $\bar{\mu}(N)$ of **(a)** Fe, **(b)** Co, and **(c)** Ni clusters as a function of the number N of cluster atoms. In view of the results in Table 1, a b.c.c– cube for Fe clusters, a f.c.c– cube for Ni clusters, and a f.c.c– octahedron for Co clusters is assumed. The *solid curves* are calculated by (4) assuming a statistical shell-by-shell growth of the clusters. Only cluster structures with completely filled atomic shells are considered. We use for Fe the values $\mu_{at} = 4.0\mu_B$, $\mu_{surf} = 3.0\mu_B$, $\mu_{bulk} = 2.21\mu_B$, for Co $\mu_{at} = 3.0\mu_B$, $\mu_{surf} = 1.9\mu_B$, $\mu_{bulk} = 1.72\mu_B$, and for Ni $\mu_{at} = 1.2\mu_B$, $\mu_{surf} = 0.7\mu_B$, $\mu_{bulk} = 0.62\mu_B$. The *dashed curves* are calculated from (5) with magnetic moments $\mu(q_i)$ dependent on the number of nearest neighbors q_i of lattice site i. We assume for Co clusters $\mu(1) \div \mu(3) = 3.3\mu_B$, $\mu(4) \div \mu(7) = 2.3\mu_B$, and $\mu(8) \div \mu(12) = 1.72\mu_B$; for Ni clusters $\mu(1) \div \mu(3) = 2.0\mu_B$, $\mu(4) \div \mu(7) = 0.9\mu_B$, and $\mu(8) \div \mu(12) = 0.62\mu_B$. For Fe clusters we put $\mu(q_i) = 4.0\mu_B$ if $q_i < 4$ and $q_i^{(2)} < 4$, $\mu(q_i) = 3.0\mu_B$ if $q_i < 4$ and $q_i^{(2)} \geq 4$, and $\mu(q_i) = 2.21\mu_B$ if $q_i \geq 4$. $q_i^{(2)}$ is the number of next nearest neighbors. The *stars* ($\star$) refer to measurements of transition metal clusters [1, 2]

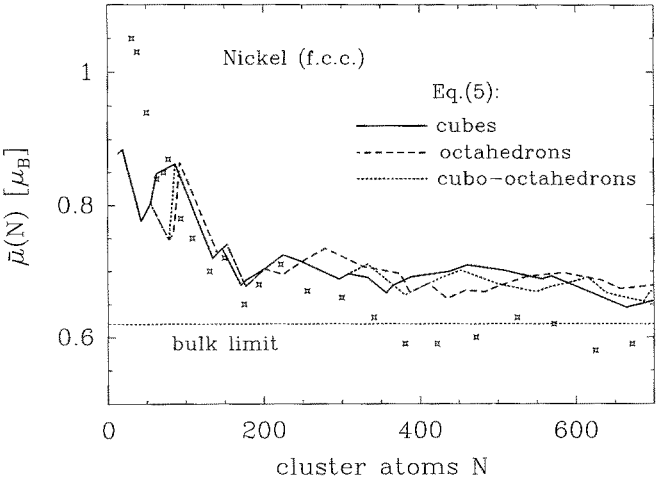

Fig. 2. Results for the magnetic shell structure of the average magnetic moment per atom $\bar{\mu}(N)$ of Ni clusters as a function of the cluster size N. A f.c.c. lattice structure with cube like, octahedral like, and cubo-octahedral like cluster shapes are assumed as indicated. $\bar{\mu}(N)$ is calculated by use of (5), assuming the same values of the magnetic moments $\mu(q_i)$ as used in Fig. 1c

Such a model has been used for describing the environmental dependence of magnetic moments in alloys [8]. When applied to clusters, q_i and μ_i are calculated for each cluster atomic site. This model takes into account best the actual atomic cluster structure. The values of μ_i used in our calculations are monotonous functions of q_i and are similar to values of magnetic moments obtained for surfaces and thin films [6]. As is known in the case of a b.c.c. lattice the atomic magnetic moment will depend also on the number of *next nearest neighbors* $q_i^{(2)}$. Thus, for clusters with a b.c.c.– structure we consider also $q_i^{(2)}$ for the determination of μ_i. The cluster atomic sites are occupied subsequently. From (5) one gets immediately the average cluster magnetic moment $\bar{\mu}(N) = (\sum_{i=1}^{N} \mu_i)/N$.

In the following we present results for the average magnetic moment per atom as a function of the cluster size N of ferromagnetically ordered clusters, corresponding to the cluster magnetization at $T = 0$. In Fig. 1 the average magnetic moment $\bar{\mu}(N)$ is given, obtained from the simple models as described by (4) or (5). The results are compared with Stern– Gerlach measurements [1, 2] for Fe, Co, and Ni clusters in the size range $30 < N < 700$. As an example, we have assumed for the atomic structure b.c.c.– cubes for Fe clusters, f.c.c.– octahedrons for Co clusters, and f.c.c.– cubes for Ni clusters, as suggested in Table 1 from the minima of $\bar{\mu}(N)$. The average magnetic moment calculated by the statistical model, (4), yields the observed minima of $\bar{\mu}(N)$. The more sophisticated model, (5), improves the magnitude of $\bar{\mu}(N)$, whereas the difference between the different cluster structures is less pronounced. This is demostrated in Fig. 2 for $\bar{\mu}(N)$ of Ni. Considering clusters shapes corresponding to cubes, octahedrons, and cubo-octahedrons, one gets an idea how much $\bar{\mu}(N)$ depends on the cluster structure. The agreement with experiment is satisfactory in view of the simple theoretical models. However, a clear evidence for a distinct cluster structure cannot be extracted from comparison with the available experimental data of the cluster magnetization.

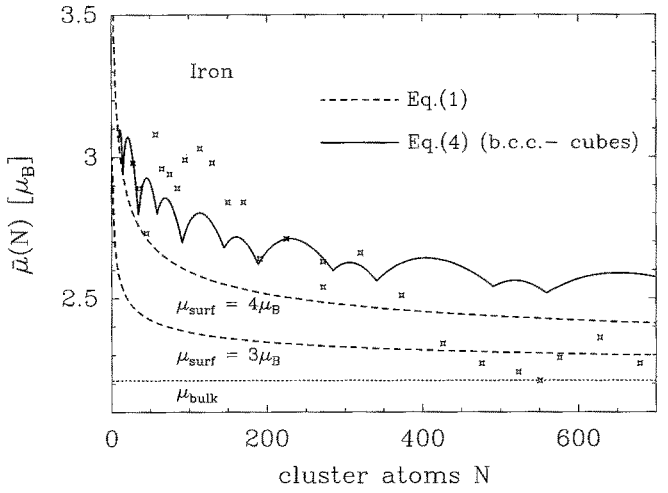

Fig. 3. Average magnetic moment per atom $\bar{\mu}(N)$ of Fe clusters as a function of the cluster size N. The dashed lines are calculated using (1), assuming $\mu_{surf} = 3\mu_B$ and $\mu_{surf} = 4\mu_B$ of the surface magnetic moment, and $\mu_{bulk} = 2.21\mu_B$. In addition $\bar{\mu}(N)$ is shown using (4), considering cluster shapes refering to rounded cubes as well as to cubes with filled shells (*full line*), s. Table 1, in contrast to the full line of Fig. 1a. The same values of the magnetic moments are taken as in Fig. 1a

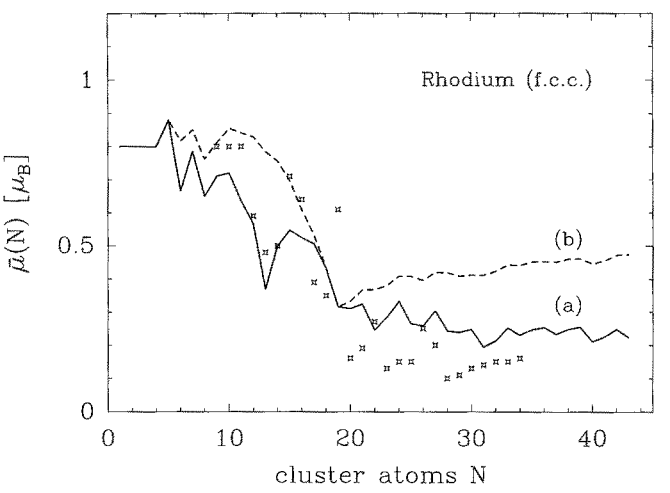

Fig. 4. Magnetic shell structure of Rh clusters with a cap– like growth of a f.c.c.– cubo-octahedron as a function of the number of cluster atoms N. We use (5) and assume $\mu(3) = 0.8\mu_B$, $\mu(4) = 1.0\mu_B$, $\mu(6) = 0.7\mu_B$, and $\mu(q) = 0$ for $q \geq 7$. Furthermore, we put (*a*) $\mu(5) = 0.4\mu_B$ (*solid line*), and (*b*) $\mu(5) = 0.85\mu_B$ (*dashed line*). The experimental results ($\star$) were obtained from [3]

In Fig. 3 we present results for Fe clusters indicating the resulting changes of $\bar{\mu}(N)$ if we perform the calculations using (4) also for rounded b.c.c.– cube clusters. Note that such a growth mode yields more minima for $\bar{\mu}(N)$. For comparison, we show also results using (1), which gives no atomic shell structure. As in Fig. 1a, the calculated results for $\bar{\mu}(N)$, except for the dashed curve using $\mu_{surf} = 3\mu_B$, are larger than the experimental ones for large clusters having more than 400 atoms. The reason for this discrepancy is not clear presently, also not for the oscillations of $\bar{\mu}(N)$ around μ_{bulk} (s. also [2] for an explanation). Possibly, (4) underestimates the increasing itineracy or cluster surface relaxation. Actually, putting $\bar{\mu}_0 = \mu_{surf}$ and $\bar{\mu}_1 = \mu_{bulk}$ as physically expected due to relaxation and neglect of surface roughness, we would get $\bar{\mu}(N) \approx \mu_{bulk}$ for the larger clusters. Regarding the comparison with experimental data, note that our results refer to $T = 0$, while the experiment observes the magnetization at $T > 0$. The experimental data are corrected to $T = 0$ afterwards by assuming the same cluster temperature for all cluster sizes. In principle, non-equilibrium effects due to increasing lattice anisotropy may cause a reduced cluster magnetization, if the relaxation time τ_N exceeds the flight time $\tau_{St.G.}$ of the clusters in the Stern– Gerlach– magnet [9]. The discrepancy between theory and experiment regarding $\bar{\mu}(N)$ calculated by (4) of the smaller clusters with $N \lesssim 200$ results from not properly taking into account the atomic structure. This follows also from comparing with results obtained from using (5), s. Fig. 1a.

In Fig. 4, we present results for the cluster size dependent magnetic moment of small Rh clusters ($N \leq 43$). Quite interestingly, these clusters are found to order ferromagnetically [3, 5], whereas Rh bulk is nonmagnetic. The values used for $\mu(q)$ are in accordance with recent first principle electronic calculations for Rh films on Ag [10], which indicate a maximum of $\mu(q)$ for $q = 4$. Concerning the cluster growth we find that growth by 'caps' rather than by a symmetrical occupation of sites in the topmost shell yields better agreement with experiment [3]. A 'cap'– like growth is expected to have a larger cohesive energy. It is to be emphasized that these results are only qualitatively valid, since the atomic magnetic moments μ_i of such small clusters should depend not only on the coordination number q_i, but also on the cluster size N and the actual cluster geometry.

In summary, the magnetization of clusters reveals interesting information regarding the interplay of atomic structure and magnetic properties. The observed oscillations of the average cluster magnet moment $\bar{\mu}(N)$ should result from magnetic shell structure and are expected to be a fingerprint of the atomic shell structure. We have proposed a model for the size dependence of $\bar{\mu}(N)$ by assuming clusters which grow shell-by-shell. The atomic magnetic moments $\mu_i = \mu(q_i)$ are assumed to depend only on their local environment, and in particular to decrease monotonous with increasing number of nearest neighbors. This reflects the increasing degree of itineracy of the atomic magnetic moments with increasing cluster size. A smooth transition to the bulk magnetic properties results. The assumption of a local environmental dependence of the atomic magnetic moments is not so valid for very small clusters, since for such systems the magnetic moments depend sensitively on the overall cluster size and structure, i.e. $\mu_i = \mu(q_i, N, \ldots)$.

Within our simple model we find an atomic shell structure for the average cluster magnetic moment $\bar{\mu}(N)$, thus reflecting the cluster geometry. The minima of $\bar{\mu}(N)$ are expected to correspond to clusters with closed atomic shells. Changes of the local environment affect the atomic magnetic moments stronger for Fe than for Co and Ni [6]. In connection with the more open b.c.c. lattice structure this feature leads to a more pronounced variation of $\bar{\mu}(N)$ for Fe. It is remarkable that our simple approach yields an oscillatory structure in $\bar{\mu}(N)$ which magnitude compares well with experiment. Comparison of theoretical results with experimental data indicate certain geometrical structures for the transition metal clusters in the considered size range.

However, from the available magnetic measurements clear evidence for a distinct cluster atomic structure on the basis of our simple models cannot be drawn. From mass abundance spectra Broyer et al. concluded that Ni and Co clusters in the size range $50 < N < 800$ may be represented by an icosahedral growth mode [11]. Such an icosahedral structure is not a part of a f.c.c. or a b.c.c. lattice, but corresponds closely to a cubo-octahedral structure. As already mentioned, from the magnetic data one cannot decide that the Ni and Co clusters build up cubo-octahedrons. For Fe clusters complex structures are observed, possibly reflecting thermal disorder. Already a fair agreement with experimental results is obtained for Rh, which magnetism is believed to depend very sensitively on the atomic structure. While the interdependence of magnetic and atomic structure is of considerable interest, this remains a difficult problem due to experimental and theoretical problems. Note that also a spin dependent electronic shell structure due to density of states oscillations $\Delta n_\sigma(\epsilon)$ may be present in magnetic clusters [12]. Thus, first principle calculations as well as further measurements of the cluster magnetization are necessary for a proper analysis of the cluster geometry. Finally, we remark again that the interdependence of magnetism and atomic structure is a very important problem in particular for very small clusters deposited on a substrate, for example. A better theory may be able to determine the difference between growth by faces or shells.

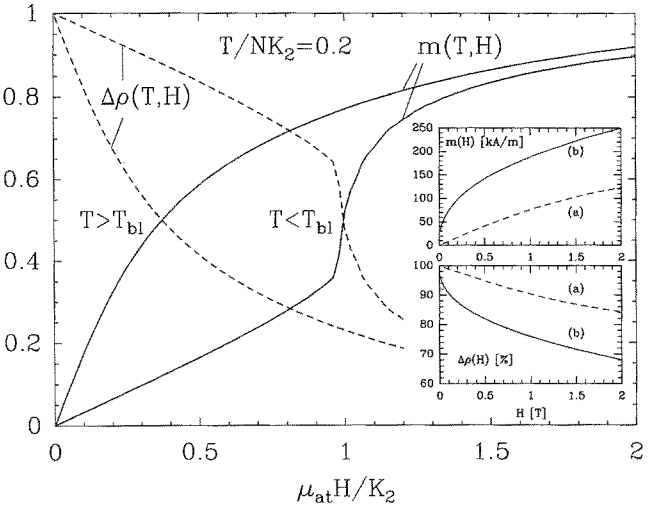

Fig. 5. Magnetization $m(T, H)$ and the corresponding giant magnetoresistance $\Delta\rho(T, H) = (\rho(T, H) - \rho(T, \infty))/(\rho(T, 0) - \rho(T, \infty))$ as a function of the external magnetic field H of an ensemble of clusters with a random orientation of the anisotropy easy axes. The cluster temperature is chosen to be $T = 0.2\, NK_2$, where K_2 is the anisotropy energy. The magnetization is calculated within statistical equilibrium ($T \gg T_{bl}$) and for strong blocking ($T \ll T_{bl}$), where T_{bl} is the blocking temperature. We assume $\Delta\rho(T, H) \propto 1 - m(T, H)$, and metallic contact between the magnetic clusters. The *inset* indicates measurements of $m(H)$ and $\Delta\rho(H)$ for layered perovskites for differently treated samples: (*a*) as deposited, and (*b*) annealed [15]

3 Magnetoresistance of a cluster ensemble

Regarding other magnetic properties of clusters, note that an ensemble of free clusters in a beam, of clusters dissolved in a metal matrix, or of deposited clusters on a surface, may in general be treated as a superparamagnetic system. For temperatures much smaller than the internal cluster ordering temperature, the clusters act magnetically like atoms with a magnetic moment $N\bar{\mu}(N)$ due to the strong (Heisenberg) exchange coupling. For such clusters the magnetic anisotropy, which is typically enhanced at surfaces or interfaces, is of particular interest with regards to many applications. This quantity determines the direction of magnetization (easy axis) within the cluster. The magnetic anisotropy can be measured with the help of the magnetization or the susceptibility of clusters in a matrix [4, 13], yielding clear deviations from the superparamagnetic (Langevin) behavior. However, the magnetic anisotropy is generally difficult to determine. In the following we discuss how this quantity may be obtained by measuring the giant magnetoresistance (GMR) of a cluster ensemble [14]. GMR originates from spin dependent electron scattering. Magnetic clusters act like paramagnetic impurities with a large magnetic moment in metallic alloys, contributing to the magnetoresistance. This was recently observed for magnetic metal clusters dissolved in a nonmagnetic metal matrix or deposited in metallic contact on a surface [7].

Consider now an ensemble of metal clusters with giant magnetic moments $N\bar{\mu}(N)$ dissolved in a metallic matrix. Magnetic interactions between the clusters are neglected first. Furthermore, an uniaxial lattice anisotropy NK_2 is present causing a random orientation of the easy axes of the clusters. Thus, for $H = 0$ the clusters represent a superparamagnetic system with a vanishing global magnetization. If an applied magnetic field H starts to align the magnetic clusters, the magnetoresistance $\Delta\rho(T, H) = (\rho(T, H) - \rho(T, \infty))/(\rho(T, 0) - \rho(T, \infty))$ decreases, and $\partial\Delta\rho(H)/\partial H < 0$. Finally, $\Delta\rho(H) \to 0$ for $H \to H_c$, when parallel orientation of the magnetic moments of all the clusters is reached. Then the system exhibits the common magnetoresistance. Thus, the behavior of GMR reflects the transition from random to ordered cluster magnetic moments. Clearly, this depends on the magnetization $m(T, H)$ of the cluster ensemble. We assume a linear dependence $\Delta\rho(T, H) \propto 1 - m(T, H)$, as suggested by experiments [7, 15]. The equilibrium magnetization $m(T, H)$ of the cluster ensemble in presence of a lattice anisotropy is similar to the Langevin function [4, 13]. However, for low cluster temperatures T such that $T \ll T_{bl} \propto NK_2$, where T_{bl} is the blocking temperature, blocking effects may be visible [16], causing a reduced magnetization of the cluster ensemble with respect to the equilibrium value. For $H\mu_{at} > K_2$ the magnetic field will overcome the anisotropy field, leading to a sudden increase of the magnetization. Thus, in view of the assumed dependence of $\Delta\rho(T, H)$ on $m(T, H)$, the GMR will exhibit a kink at $H_c = K_2/\mu_{at}$, as shown in Fig. 5. By increasing the temperature the sharp kink of the magnetization at $H\mu_{at} = K_2$ is smeared out, yielding a corresponding change of $\Delta\rho(T, H)$. Therefore, from GMR one can in principle determine the magnetic anisotropy and also the blocking temperature. We emphasize that the enhanced GMR for blocked cluster magnetic moments originates from the *random* orientation of the easy axes. In contrast, if the easy axes are aligned parallel to the magnetic field, then no GMR will result [7].

Summarizing, we have discussed the magnetoresistance of a magnetic cluster ensemble and in particular the relationship between the magnetization and the GMR of a cluster ensemble. Measurements of $\Delta\rho(T, H)$ should reflect the magnetic properties of the cluster system and may be used to determine the lattice anisotropy. If the ferromagnetic exchange interaction between the clusters dominates the lattice anisotropy, then one expects $\Delta\rho(H) \approx 0$ for the cluster ensemble. Similarly, if the magnetic dipolar interaction, which favors a ferromagnetic spin alignment parallel to the surface, is important (as in the case of magnetic clusters deposited on a metallic substrate), then also $\Delta\rho(H) \to 0$ is expected. Considering technological application of the GMR, note that a cluster system dissolved in a matrix seems to be much easier to fabricate than multilayer systems with ultrathin atomic layers.

It is interesting to remark that the magnetic properties and the enhanced magnetic anisotropy can also be determined with the help of the nonlinear magneto-optical Kerr effect [17]. Therefore, such measurements should be performed in addition to Stern– Gerlach experiments and magnetoresistance measurements to determine the magnetic properties of clusters.

Note added: After completion of this work we became aware of another model for the shell structure of the average cluster magnetic moment proposed by Billas et al. [2].

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