Phenomenological considerations in Percolating Magnetic Nanostructures

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Transport and magnetotransport properties were analysized systematically in percolating magnetic nanostructures such as Ni-rich $NiFe-SiO_2$ and $Fe-SiO_2$ films, where universal features unique to percolating magnetic nanostructures exhibiting giant Hall effect are pointed out. Assuming the presence of nanometer sized particles in the percolating conduction channels whose contribution to the conduction is sensitive to the temperature and external magnetic field, a phenomenological model was developed which explains the essence of these universal features.

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Spin-dependent transports have attracted a lot of attention since the discovery of giant magnetoresistance (GMR) in metallic multilayers like Fe/Cr or Co/Cu [1–4]. It is widely believed that the GMR in such conducting systems are results of spin-dependent scattering from magnetically heterogeneous regions where local magnetizations are antiferromagnetically arranged [5,6]. Observation of GMR in immiscible magnetic granular composite films with a metallic matrix, such as $Fe-Ag$ or $Co-Ag$ [7,8], and those with an insulating matrix, such as $Co-Al_2O_3$ [9], indicate that spin-dependent scattering or tunnelling [10] is generic in all inhomogeneous magnetic nanostructures.

The recently observed giant Hall effect (GHE) provides a new magnetic nanostructure to study spin-dependent transports with magnetic particles forming percolation conducting channels. GHE refers to a 10^4-fold enhancement in Hall resistivity in ferromagnetic granular $(NiFe)_x-(SiO_2)_{1-x}$ films for x near x_c, the percolation metal volume fraction [11], compared to that of a homogeneous ferromagnetic metal. The effect cannot be explained *via* a simple percolation theory that predicts a maximum enhancement factor of ~ 20 [12] based on the percolation critical exponent of Hall resistivity $g \sim 0.4$ [13] for a 1 μm thick film. A variety of models have also been proposed to understand the giant Hall effect [15,17,18] but are so far unsuccessful. In fact, a complete microscopic theory for the GHE seems to be beyond our reach at present moment since even in homogeneous transition metals the understanding of the extraordinary Hall effect is very limited [19,20]. The percolating labyrinth structure [12,21] adds further complication to the problem. On the other hand, the recently studied granular $Fe-SiO_2$ films [22] seems to indicate that there exists *unique* and *universal* features associated with a new conduction mechanism and spin dependent transports in magnetic metal-insulating nanocomposite films exhibiting giant Hall effect. In this paper, a systematic comparison of transport and magnetotransport properties in both the $(NiFe)_x-(SiO_2)_{1-x}$ and $Fe_x-(SiO_2)_{1-x}$ films are carried out where features universal to the two percolating magnetic nanostructures closed to the percolation volume fraction $x \to x_c$ are presented. To understand these features a phenomenological model is developed which captures the essence of these universal features and which, we believe, also provides insights in the origin of the GHE.

It was discovered that for all samples exhibiting the giant Hall effect, including both $NiFe-SiO_2$ and $Fe-SiO_2$ films which have carriers of different sign [22], there always exist a large resistivity with a $\rho(T) \sim -log(T)$-like temperature dependence. Shown in Fig. 1 is the temperature dependences of the resistivity (normalized at $5K$) plotted in a logarithmic scale for $Fe_{0.53}-(SiO_2)_{0.47}$ under different annealing conditions. For annealing below $300°C$, where giant Hall effect exists ($\rho_{xys} \sim 0.1m\Omega cm, \rho \sim 0.1\Omega cm$), the temperature dependent resistivity shows clearly a $\sim log(T)$ like dependence. Whereas, for annealing above $300°C$, where giant Hall effect disappears ($\rho_{xys} \sim 0.001m\Omega cm$), the temperature dependences become metallic with considerably lower resistivity ($\rho \sim 0.1m\Omega cm$). The same qualitative feature was found in $NiFe-SiO_2$ films [12].

We have also studied magnetoresistivity of these samples in geometries with magnetic field in the film plane and are longitudinal and transverse to the current [23]. All geometries yield about the same negative value for films near the percolation threshold, both for $NiFe-SiO_2$ and $Fe-SiO_2$ systems, indicating that the magnetoresistance is a result of spin-dependent transport processes similar to many inhomogeneous magnetic nanostructures where giant magnetoresistance were observed [1–9]. To investigate further the spin dependent transport mechanism of the magnetoresistance in the region of giant Hall effect, we plot in Fig. 2 the magnetoresistivity $-\Delta\rho$ versus resistivity in a log-log scale for Ni-rich $NiFe-SiO_2$ (circle) and $Fe-SiO_2$ (square) samples measured at $300K$. Note that the value of $-\Delta\rho$ in $Fe-SiO_2$ was divided by 4 to take into account the difference in the saturation magnetization because $-\Delta\rho$ scales with M^2 for a spin-dependent process. Solid line

in this figure is the fitting result according to $-\Delta\rho \sim \rho^b$, with $b \sim 1.2$. It is clear that within experimental error the same power law is found for both systems, indicating that the same spin-dependent transport mechanism is functioning in both systems exhibiting the giant Hall effect, irrespective to the sign of the carriers and the details of the microstructures.

We propose that the universal behaviours associated with GHE observed in two systems with very different microscopic characters are results of their common percolating, granular structure. To understand these universal behaviours we first consider a small metallic particle of size s^3 in a percolating conducting channel and ask the question: under what condition the particle can be considered as conducting? In general a metallic particle can be considered as conducting if there is no energy barrier against transporting electrons in and out of the particle. This is the case if the thermal energy $\sim k_BT$ of electrons is larger than the energy level spacing inside the particle, $\Delta E(s) \sim \frac{\hbar^2}{2m^*s^2}$, where m^* is the electron effective mass, or $\Delta E(s) \sim \frac{e^2}{\epsilon s}$, where ϵ is the effective dielectric constant, if Coulomb blockade effect is important [24]. In our granular systems where it was found that large number of small size particles $\leq$ 3nm exist [12], it is expected that this finite size effect will be important. The small particles which behave as metals at high temperature, would become insulating at temperature $k_BT \leq \Delta E(s)$, and the effective conducting volume fraction decreases with temperature, leading to increasing resistance at low temperature. In the case when the particles are magnetized and with magnetization pointing in random directions, there is an additional energy barrier between particles with magnetization pointing in opposite direction, and these particles become insulating with respect to each other. This energy barrier is removed when the magnetization of the particles are aligned, as is in the case when a large magnetic field is applied, leading to an increase in effective volumn fraction and negative magnetoresistance. To capture these effects at least crudely we assume that the transport properties of the magnetic granular systems under consideration can be described by effective classical percolation models near percolation threshold, where the resistivity is given by the formula

$$\rho(T,H) = \frac{\rho_0}{(x_{eff}(T,H) - x_c)^t}, \qquad (1)$$

where ρ_0 is a material dependent parameter and t is a critical exponent characterizing the effective classical percolation model, $x_{eff}(T,H)$ is the effective conducting volume fraction. H is the external magnetic field. In particular, we expect from our previous discussion that at zero external magnetic field,

$$x_{eff}(T,0) \sim \int_{s(T)}^{\infty} (s^3)n(s)ds = x_0 - \int_0^{s(T)} (s^3)n(s)ds, \qquad (2)$$

where $n(s)ds$ is the number of particles in the sample with linear dimension between s and $s+ds$, $x_0 = \int_0^{\infty}(s^3)n(s)ds$ is the metal volume fraction, which was determined by EDX. $s(T)$ is a size cutoff determined by the equation $k_BT = \Delta E(s)$. At finite magnetic field, we expect that x_{eff} increases because of the alignment of magnetization between different particles. The precise magnetic field dependence of this effect is hard to estimate. However, at $H > H_s$ where magnetization saturates, the magnetic field dependence should vanish and we expect

$$x_{eff}(T, H > H_s) \sim x_{eff}(T,0) + \Delta x_M(T), \qquad (3)$$

where $\Delta x_M(T) \sim A\int_{s(T)}^{\infty}(\lambda)(s^2)n(s)ds$, and A is an unknown constant depending on the microscopic details of the material, $\lambda \sim$ thickness of the magnetic domain wall separating particles with opposite magnetization orientation [25].

Substituting Eq. (2) in Eq. (1), and assuming that the change in x_{eff} is small, we obtain at zero magnetic field,

$$\rho(T,0) \sim \rho_\infty \left(1 + t(\frac{\rho_\infty}{\rho_0})^{\frac{1}{t}} \int_0^{s(T)} (s^3)n(s)ds\right), \qquad (4)$$

where $\rho_\infty = \rho_0/(x_0 - x_c)^t$ is the high temperature resistivity. In particular,

$$\frac{d\rho(T,0)}{dT} = t\rho_\infty(\frac{\rho_\infty}{\rho_0})^{\frac{1}{t}} \frac{ds(T)}{dT}(s(T))^3 n(s(T)), \qquad (5)$$

which provides a precise relation between $d\rho(T,0)/dT$ and particle size distribution $n(s(T))$ which can be tested experimentally. In particular, at the temperature region where $\rho(T,0) \sim -log(T)$, we find correspondingly $n(s) \sim s^{-4}$ [26]. Notice that this behaviour is independent of the orgin of $\Delta E(s)$ and stays the same as long as $\Delta E(s) \sim s^{-a}$, with $a \geq 1$. Rapid increase in $n(s)$ as s decreases was indeed observed experimentally [12] for samples with $\rho(T) \sim -log(T)$. For annealed samples where the $\rho(T) \sim -log(T)$ behaviour is absent, it was also observed that the rapid increase in $n(s)$ at small s is destroyed and the peak in $n(s)$ was gradually shifted to higher values of s when annealing temperature is increased [12]. The same qualitative behaviour was also obtained from Eq. (5) using the experimental data for $\rho(T,0)$ as input. Figure 3 shows the particle size distribution $n(s)$ for $(NiFe)_x - (SiO_2)_{1-x}$ films (a) as deposited, and (b) annealed as $520°C$. The solid lines are results of TEM measurement reproduced from Ref. [12], whereas dash-lines are results obtained from the corresponding resistivity data using Eq. (5) with $\Delta E(s) = \frac{\hbar^2}{2ms^2}$, where m is the free electron mass. The qualitative agreement between theory and experiment is apparent. Fitting with $\Delta E(s) = e^2/\epsilon s$ to experimental data was also tried but the agreement with experimental data is slightly worse. However, it must be cautioned that so far the available experimental data and the theory are both too crude

to determine quantitatively what is the precise orgin of $\Delta E(s)$ in our systems. Using Eqs. (1) and (3), it is also easy to show that the saturated magnetoresistivity $\Delta\rho$ is given by

$$\Delta\rho = -t\rho_\infty \left(\frac{\rho_\infty}{\rho_0}\right)^{\frac{1}{t}} \Delta x_M(T). \quad (6)$$

Using $300K$ as the high temperature limit, we obtain $\Delta\rho \sim (\rho_\infty)^b \sim \rho^b$ near the percolation threshold, where $b = 1 + t^{-1}$, which, within experimental accuracy, is consistent with experimental values of $b \sim 1.2 \pm 0.1$ and $t \sim 3.6 \pm 1.0$ [11]. It is also easy to see from Eq. (4) that our phenomenological theory also predicts that the temperature dependent part of the resistivity at $H = 0$ should also scale similarly as $\Delta\rho$, i.e. $\Delta\rho' = \rho(T,0) - \rho_\infty \sim (\rho_\infty)^{1+t^{-1}}$. Taking $T = 77K$, we show in the upper panel of Fig. 2 the temperature dependent part of resistivity $\Delta\rho'$ versus resistivity $\rho(300K)$ in a log-log scale for $(NiFe)_x - (SiO_2)_{1-x}$ samples (filled circles). Here only samples with $\rho \sim 0.4\Omega cm$ or above were shown, because lower resistivity samples have minimum in the temperature dependence. The similar scaling behaviour of $\Delta\rho$ and $\Delta\rho'$ versus ρ is apparent from the figure, irrespective of the crudeness of our approach.

It has to be emphasized that the temperature dependence of resistivity and magnetoresistance are usually results of very different physical processes and it is highly unusual that they both scale with resisitivity with a similar critical exponent. The observed rough agreement in the critical exponents of the two resistivities strongly supports our physical picture of temperature and magnetic field dependent effective conduction volume fraction which dominates the physics of percolating magnetic nanostructures.

Note that our phenomenology in describing the dependencies of resistivity on the temperature, the magnetic field, and the particle size distribution, in various percolating magnetic nanostructures, is reasonably successful. This leads us to ask the question of whether similar phenomenology can be applied to describe the Hall resistances, and in particular, whether the physical origin of the GHE can be identified from our phenomenology. Following our previous analysis, we propose that the Hall resistivity can be described by the formula

$$\rho_{xy}(T,H) = \frac{BR_0}{(x_{eff}(T,H) - x_c)^\xi}, \quad (7)$$

where R_0 is a material dependent parameter and $B = H + 4\pi M$ is the total magnetic field seen by electrons in the system. The Hall number $R_H \sim \rho_{xy}/H \sim (x_{eff}(T,H) - x_c)^\xi$ and extraordinary resistivity $\rho_{xys} \sim M/(x_{eff}(T,H) - x_c)^\xi$ are predicted to scale with $\rho(T,H)$ with the same exponent ($R_H(\rho_{xys}) \sim \rho^{\frac{\xi}{t}}$) near the percolation threshold in our phenomenology, which is consistent with existing data where $\xi/t \sim 0.8$ within the experimental accuracy [27]. Note that scaling between two physical quantities are much more reliable as it is insensitive to the determination of both the metallic volume fraction and its critical value separating the metal-insulator phases. This is particularly important for the percolating nanostructure where the critical value is not sharply defined as in the classical percolation system.

Perhaps the most surprising results in our phenomenology are the largeness of the values of exponents ξ and t deduced experimentally where $t \sim 3.6$ and $\xi \sim 2.9$, which are unexpected in existing theoretical framework. These values are much larger than the corresponding values obtained from usual classical percolation models [13] $t \sim 2$ and $\xi \sim 0.4$, suggesting that the effective classical percolation models describing our experimental systems are rather unusual. In particular, the large enhancement of the critical exponent ξ (a factor of 7 of theoretical value of 0.4) is responsible for the large magnitude of Hall resistance observed near the percolation threshold, whereas the relatively smaller enhancement in t (by a factor of 1.8) seems to be the reason why the enhancement in magnetoresistance is not as drastic as the Hall effect. The key to understand the Giant Hall effect seems to lie in the understanding the physical origin of these unusually large values of critical exponents.

Summarizing, in transport properties in percolating magnetic nanostructures exhibiting giant Hall effect, we have identified a few unique correlated features, namely the correlation between a -log(T) like temperature dependent resistivity and a particle size distribution having a large fraction of small nanometer sized particles, power law dependencies between the magnetoresistivity and the room temperature resistivity, and that of temperature dependent part of resistivity and room temperature resistivity. These unique features are explained by a phenomenological model developed in this work, assuming the presence of nanometer sized particles in the percolating conducting channels, whose contribution to the conduction is sensitive to the temperature and external magnetic field. We believe that the key to understand the origin of the giant Hall effect lies in the understanding of the unusually large critical exponents.

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FIG. 1. Resistivity as a function of temperature for $Fe_{0.53} - (SiO_2)_{0.47}$ for various annealing conditions.

FIG. 2. Magnetoresistivity, -$\Delta\rho$, as functions of resistivity, ρ, in a log-log plot for $NiFe - SiO_2$ (open circle) and $Fe - SiO_2$ (open square) (lower panel), and temperature dependent part of resistivity, $\Delta\rho'$, versus ρ for $NiFe - SiO_2$ (filled circle).

FIG. 3. Particle size distributions ($\times 10^{11}/mm^2$) determined by TEM (solid line) and from Eq.(5) using temperature dependent resistivity data (dash-line) for (a)$Ni_{0.55} - (SiO_2)_{0.45}$ as deposited ($\bigcirc$(TEM);∇(theory)), and (b)$Ni_{0.55} - (SiO_2)_{0.45}$ annealed at 520^oC ($\square$(TEM), Δ(theory)).

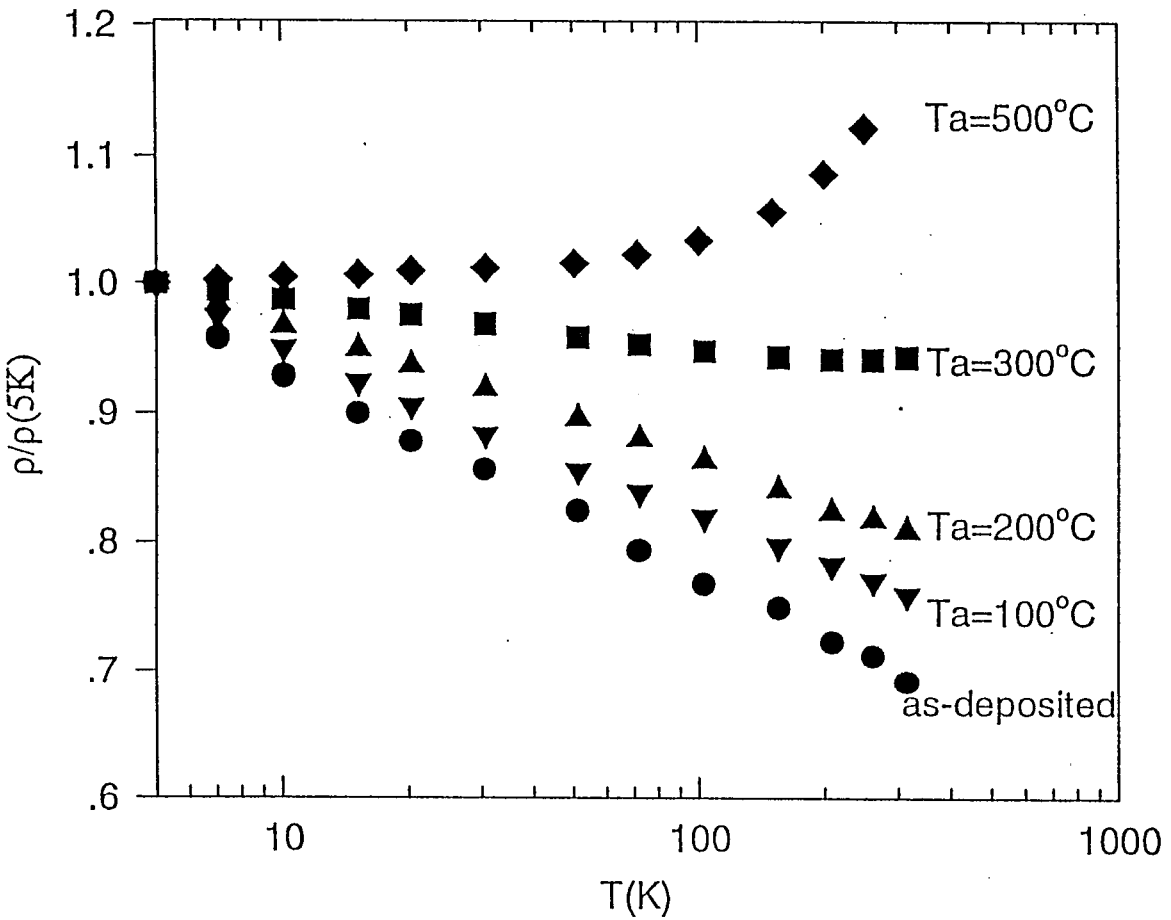
1.2
1.1
1.0
.9
.8
.7
.6
ρ/ρ(5K)
10
100
1000
T(K)
Ta=500°C
Ta=300°C
Ta=200°C
Ta=100°C
as-deposited

Fig. 1

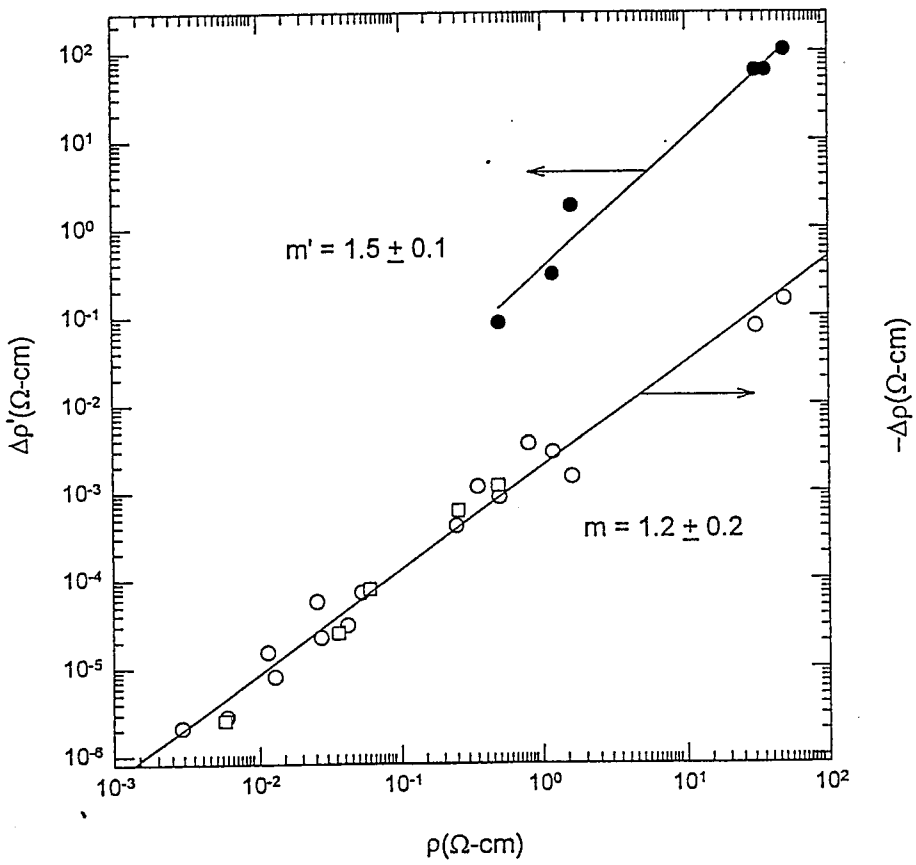
m' = 1.5 ± 0.1
m = 1.2 ± 0.2
Δρ'(Ω-cm)
−Δρ(Ω-cm)
ρ(Ω-cm)
10²
10¹
10⁰
10⁻¹
10⁻²
10⁻³
10⁻⁴
10⁻⁵
10⁻⁶
10⁻³
10⁻²
10⁻¹
10⁰
10¹
10²

Fig. 2

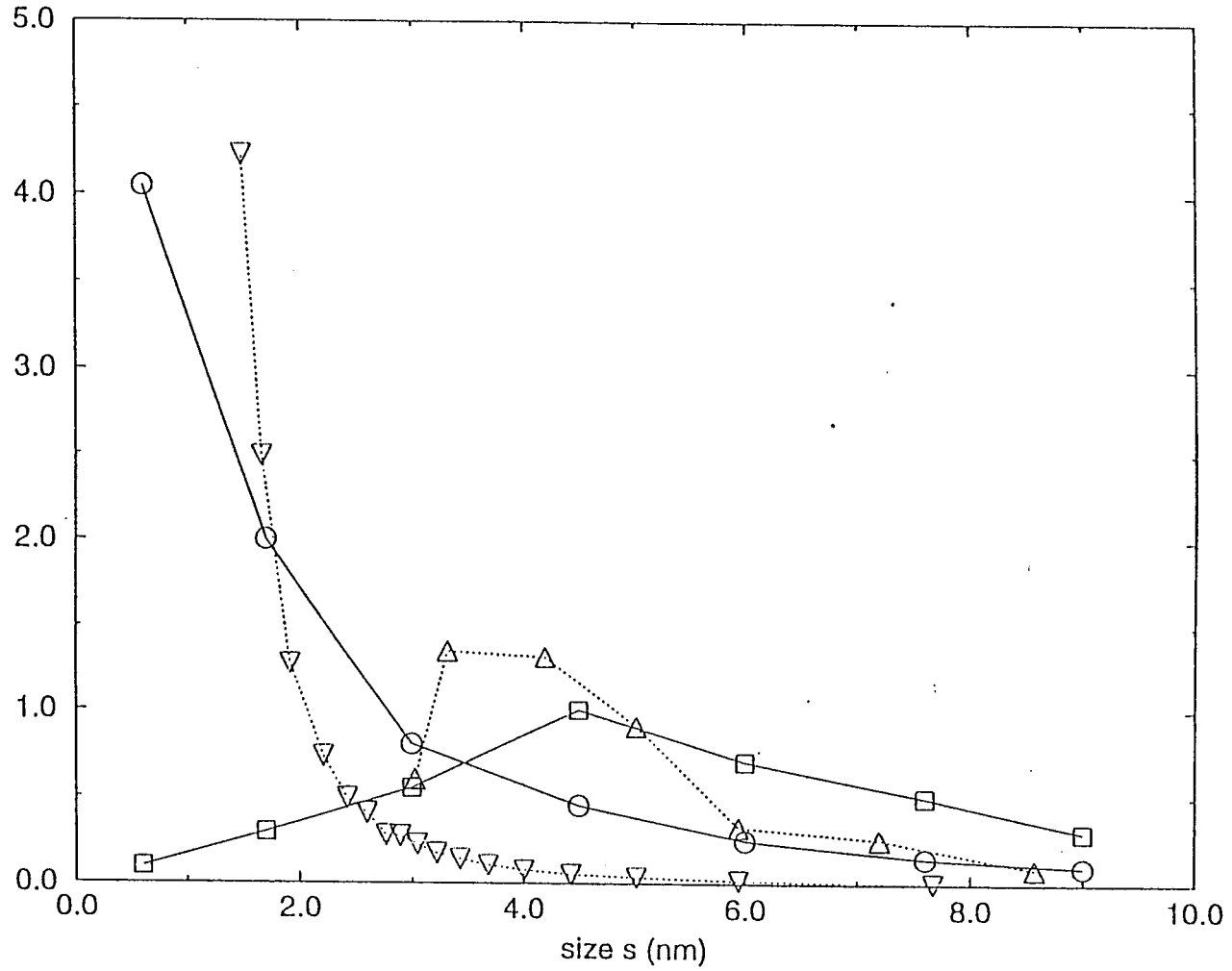

Fig. 3