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Study of relaxation of a cluster of magnetic particles in magnetic field

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A setup is described intended to study the conversion of a cluster of magnetic particles into a supercluster under magnetic field influence. The technique used makes it possible to study the processes associated with the magnetic particle motion during the supercluster formation, the contribution of the domain wall displacement inside the particles to the measured signal being negligible. The relaxation processes during a Ni supercluster formation in magnetic fields of 0 to 160 Oe strength have been studied. The presence of slight changes in the cluster state in the fields lower than the first critical one has been revealed, where no changes in the cluster geometry have been observed before. The typical relaxation curve shape has been found to depend on the formative field amplitude. The saturation time of the particle redistribution process depends non-monotonously on the field amplitude. The dependence maximum is located between the first and second critical fields.

В работе представлена установка для исследования процессов распада кластера магнитных частиц в суперкластер под воздействием магнитного поля. Используемая методика позволяет исследовать процессы, связанные с перемещением магнитных частиц при формировании суперкластера, при этом вклад в измеряемый сигнал от смещения доменных стенок внутри частиц пренебрежимо мал. Исследованы процессы релаксации при формировании суперкластеров частиц Ni при воздействии магнитного поля с напряженностью 0–160 Å. Установлено наличие слабых изменений состояния кластера в полях меньших первого критического, в которых изменений геометрических характеристик кластера ранее не наблюдалось. Установлено, что характерный вид релаксационной кривой зависит от амплитуды формирующего поля. Время выхода на насыщение процесса перераспределения частиц зависит от амплитуды поля не монотонным образом. Максимум этой зависимости расположен между первым и вторым критическим полями.

The behavior of clusters of magnetic particles in a magnetic field is studied intensely both from theoretical [1, 2] and applied [3–6] standpoint. So, in [1], the formation of ordered structures in a magnetic field was observed for clusters located at a liquid/liquid interface. In [3, 4], when studying stationary states of magnetic particle superclusters formed in the fields with increasing strength, the existence of critical fields has been revealed, these fields being responsible for the changes in the magnetic particle motion as the field strength increases. At to the applied aspect, the structures formed from the magnetic particle clusters in a magnetic field could be used as multilevel packings in high-gradient magnetic filters [4, 5].

The observations have shown that the supercluster formation processes under the formative field application are accompanied by the relaxation phenomena, the latter consisting in that the equilibrium cluster configuration is not formed instantaneously. The various relaxation aspects in magnetic fields for systems of magnetic particles (both fixed in a rigid matrix, see [6] and references therein, and free-moving [7, 8]) have been studied. When a supercluster is formed from an ensemble of magnetic particles under applied magnetic field, of a substantial importance is the formation process of individual clusters and the mo-

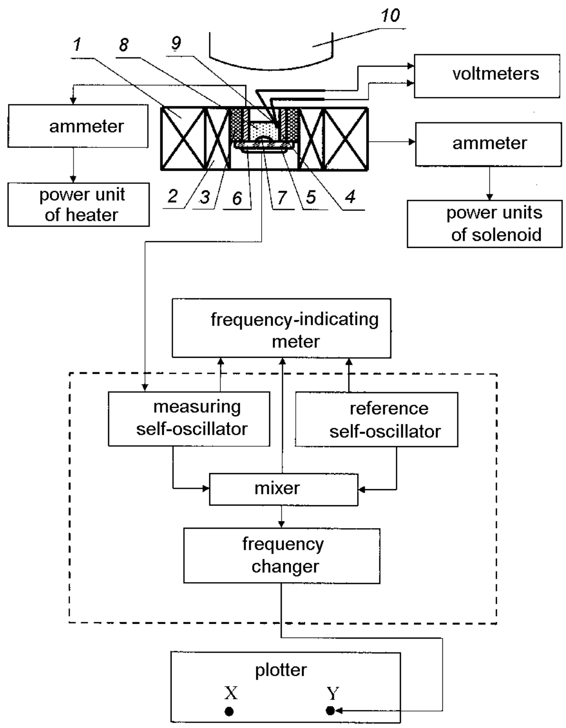

Fig. 1. Setup schematic view: *1*, *2* — magnetic systems to generate the constant and variable magnetic fields; *3* — cell; *4* — heater; *5* — sensor; *6* —glass cell bottom; *7* — cluster; *8* — paraffin; *9* — thermocouple; *10* — microscope.

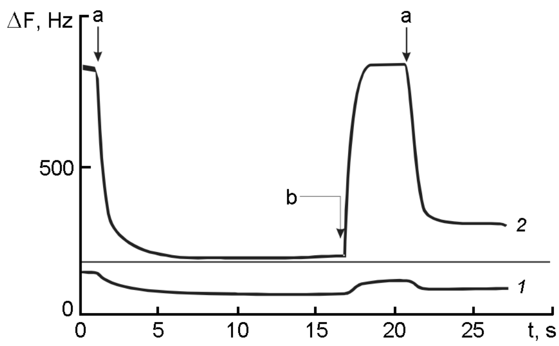

Fig. 2. Relaxation at switching on and off of a constant field at different amplitudes: a and b, the switching-on and switching-off moments. Field strength (Oe): 40 (*1*), 70 (*2*).

tion thereof relative to the initial positions and to each other during the equilibrium state establishing [4, 5]. That is why the studies of the process dynamics are necessary. The purpose of this work is to study the time dependences of the supercluster parameters during the formation thereof and to define the factors influencing the shape of those dependences.

To investigate the time characteristics for conversion of a cluster of magnetic particles into a supercluster under magnetic field action, a setup has been elaborated and constructed basing on the induction-frequency method. The setup schematic diagram is presented in Fig. 1. The main setup part is an auto-generator assembly (denoted by a dashed line in the Figure) consisting of two LC generators, the one being a reference, the other serves to measure. The scheme includes a mixer giving at the output the frequency difference $\Delta F = |f_{ref} - f_{meas}|$. The frequency-to-voltage converter transforms the ΔF into a DC voltage that comes to the Y input of a recorder.

The setup operates as follows. The initial cluster of magnetic particles (*7*) formed in molten paraffin (*8*) forms the core of the induction coil (*5*) of the measuring autogenerator loop operating at the frequency of 1 MHz. The coil (*5*) is shaped as a flat spiral fixed by a glue under the glass bottom (*6*) of a dielectric cell (*3*). The glass is 0.17 mm thick. The constant magnetic field and the variable one generated by coils (*1*) and (*2*) are applied perpendicular to the substrate plane and thus to the developed cluster surface. The action onto the cluster results in changes of its parameters and thus the measuring generator changes its frequency. The signal proportional to ΔF comes to the Y input of the recorder, the time axis being the X one. The plot $\Delta F(t)$ obtained in this manner is referred to as the relaxation curve.

The relaxation curves are recorded as follows. The initial clusters are formed by melting the feed balls made of a mixture of paraffin with nickel powder and placed in the bottom center of the cell filled with molten paraffin heated up to 100°C. The feed ball diameters are calibrated to within 10%. The initial cluster formation technique is described in detail elsewhere [4]. Then the formative magnetic field of a preset strength is applied and during the supercluster formation, a signal in proportion to ΔF is recorded as a function of time t.

Typical dependences for $\Delta F(t)$ as the constant magnetic field of different amplitudes is successively switched-on and switched-off are shown in Fig. 2. It is seen that both the field switching and cutoff result in a substantial changes in ΔF. This may be connected with changes in arrangement of the particles in the cluster, in the shape of the latter, and in the domain configurations within individual particles. The characteristic time values in the curve section following the moment marked by the arrow **a** are of the order of several seconds and exceed the duration of the magnetic field pulse increase that does not exceed 0.2 s. Thus, the section of $\Delta F(t)$ dependence following the point **a** evidences a relaxation in the system.

The visual observations during the field application have shown that, starting from the formative field strength of about 50 Oe (being the 1st critical field H_1 according to [3]), individual clusters are separated from the solid initial cluster body and move radially from the center. As the field is switched-off, these clusters fall down. Also it has been found in [3] that as the formative field exceeds the second critical value ($H_2 \approx$ 100 Oe), the particle start to move not only along the substrate surface but perpendicular thereto, along the gradient of the scattering fields of individual particles.

It is to note that the relaxation observed at the field switching-on is observable also at its successive switching-off and switching-on (Fig. 2, the section following the arrow **b**). The characteristic relaxation times at the field switching-on and switching-off are commensurable to each other.

To discriminate the contribution of the motion of the particles themselves from that of the motion of the domain walls therein in the $\Delta F(t)$ dependence, the $\Delta F(t)$ plot was recorded for the initial cluster frozen in paraffin. The solidified paraffin did not allow the magnetic particles to move, so, in this case, the ΔF changes were due only to the changes in the powder particle domain state. The typical dependences are shown in Fig. 3a, b. In both cases, the 80 Oe field was applied to the initial cluster formed in zero field. Comparison of the dependences shown in Fig. 3 suggests that it is just the changes in the particle positions with regard to the measuring sensor that contribute mainly to the ΔF changes. Thus, the $\Delta F(t)$ dependences obtained later at T = 100°C can be suggested to be due mainly by the changes in the particle arrangement.

It has been shown before [3, 4] that the particle motion run at the initial cluster conversion in various formative fields can be subdivided into three characteristic regions corresponding to the field values lower than the 1st critical one (H_1), those between the 1st and 2nd (H_2) critical fields, and above the 2nd critical field. Therefore, the effect of the field strength on the relaxation curve was studied. For initial clus-

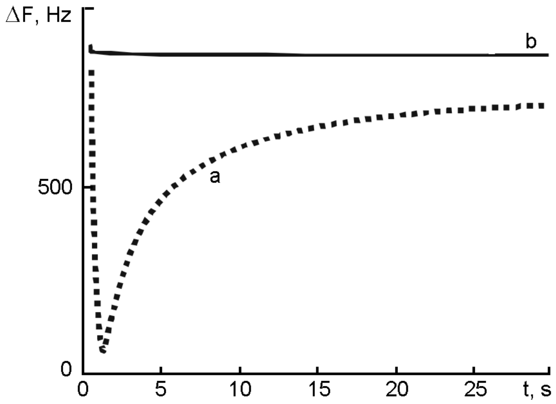

Fig. 3. Comparison of time dependences at the magnetic field action onto a cluster with mobile particles **(a)** and those frozen in paraffin **(b)**.

ters formed from the feed balls of 1.0±0.1 mm in diameter, the $\Delta F(t)$ dependences were recorded within the magnetic field range of 0 to 160 Oe covering the three characteristic regions. The typical $\Delta F(t)$ dependences recorded during the cluster conversion in constant magnetic fields of different strength are shown in Fig. 4. It is seen from the Figure that changes in the magnetic field pulse amplitude results in substantial differences between the $\Delta F(t)$ curve shapes. The $\Delta F(t)$ curves recorded in fields up to 80 Oe show a monotonous time dependence of ΔF. Exceeding of the above field value results in a non-monotonous $\Delta F(t)$ run. Consideration of $\Delta F(t)$ dependences at different constant field amplitudes shows that a characteristic valley occurs in the field interval between H_1 and H_2.

Moreover, it is to note a nonzero ΔF changes in low fields of the order of 30 Oe that evidences the cluster particle motion in fields lower than H_1 where no changes in the cluster geometric parameters is observed. Note that the $\Delta F(t)$ change at the conversion of a cluster of free particles in the 30 Oe field exceeds that occurring under the action of the 80 Oe field onto the body of frozen particles. Thus, the technique used makes it possible to record the particle position changes in the fields lower than the 1st critical one (Fig. 4, the section **a**).

The combined consideration of the dependences recorded within the whole magnetic field range shows that the formative field value affects substantially the relaxation curve parameters R, P, T1, and T2 (notation see Fig. 4). In Fig. 4, T2 refers to the time interval where the $\Delta F(t)$ change rate drops down to 20 Hz/s (the criterion of

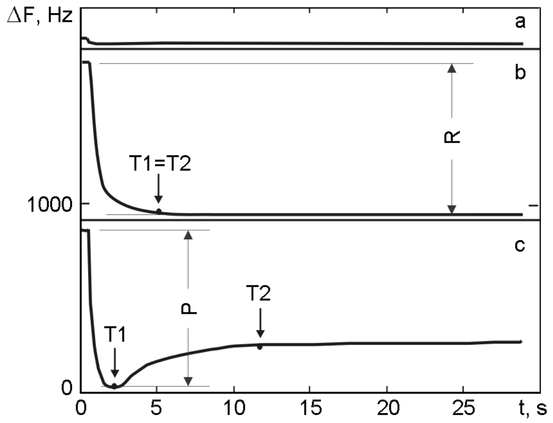

Fig. 4. Time dependences of the measuring generator frequency at the conversion (decay) of the clusters of magnetic particles at different amplitudes of constant field. Field strength (Oe): 30 (a), 60 (b), 130 (c).

the process saturation). T1 denotes the time moment where the dependence minimum is attained.

Fig. 5 shows the characteristic time values T1 and T2 as the functions of the formative field amplitude. In the Figure, the vertical line pairs I and II denote the regions including the critical fields H_1 and H_2. These dependences are seen to be non-monotonous and to have a maximum in the interval between the 1st and 2nd critical field values, thus evidencing a change in the motion character of the cluster particles within that interval.

Note that in the 160 Oe field, the time to the T1 minimum becomes comparable to the magnetic field increase time at switching-on thereof, while the total duration of the cluster conversion, T2, exceeds considerably the field increase one. The T2 shortening in strong fields (curve 2) indicates that as the constant magnetic field amplitude rises, the particle interaction forces increase substantially, therefore, the general motion character in the system of magnetic particles is established more rapidly.

To conclude, the technique developed makes it possible to study the phenomena associated with motion of magnetic particles in the course of a supercluster formation. The displacement of the intraparticle domain walls contributes only negligibly to the signal measured. The supercluster formation at the pulse magnetic field switching-on is accompanied by the relaxation processes. Similar relaxation occurs as the formed supercluster is subjected to pulses of the switched-on and switched-off magnetic field causing the orientation changes of individual clusters. Slight changes in the cluster state are observed in the fields lower than the 1st critical one, where no changes in the cluster geometric parameters were observed before. The relaxation curve characteristic shape depends on the formative field amplitude. The saturation time (T2) of the particle redistribution process depends non-monotonously on the field amplitude. The dependence maximum is localized between the 1st and 2nd critical field values. The measurement of the ferromagnetic response characteristics can be used top study the internal rearrangement of individual clusters during the supercluster transformations and makes it possible to study the characteristic time intervals defining the processes running therein.

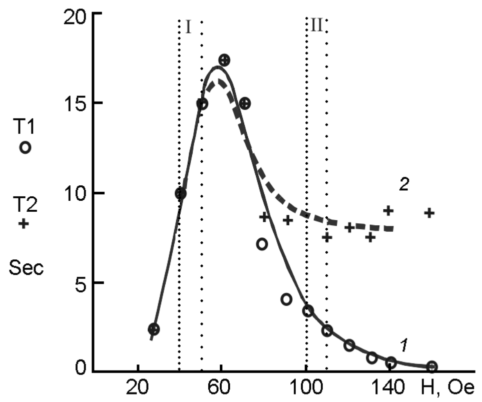

Fig. 5. Characteristic times as functions of the formative field amplitudes.

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Дослідження релаксації кластера магнітних часток у магнітному полі

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Представлено устаткування для дослідження процесів розпаду кластера магнітних часток у суперкластер під дією магнітного поля. Методика дозволяє досліджувати процеси, які пов'язані з переміщенням магнітних часток при формуванні суперкластера, при цьому вклад у сигнал, який вимірюється, від зміщення доменних стінок у частинках достатньо малий. Досліджено процеси релаксації при формуванні суперкластерів частинок Ni при дії магнітного поля з напруженістю в інтервалі 0–160 Å. Встановлено наявність змін стану кластера у полях менших першого критичного, в яких змін геометричних характеристик кластера раніше не спостерігалось. Встановлено, що характерний вид релаксаційної кривої залежить від амплітуди формуючого поля. Залежність від амплітуди поля часу виходу на насичення процесу перерозподілу часток не монотонна. Максимум залежності лежить між першим та другим критичними полями.