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Anomalously large magnetostriction caused by rearrangement of magnetoelastic domains

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Anomalous large values of magnetostriction related to rearrangement of multidomain state in easy-plane antiferromagnetics of iron group dihalides have been shown to be associated with magnetoelastic nature of the multidomain state. To describe that multidomain state, a parameter has been introduced which characterizes the average orientation of domains. The parameter is referred to as the domain co-alignment one.

Показано, что аномально большие величины магнитострикции при перестройке многодоменного состояния легкоплоскостных антиферромагнетиков дигалидов группы железа связаны с магнитоупругой природой их многодоменного состояния. Для описания такого многодоменного состояния введен параметр, характеризующий среднюю ориентацию доменов, который обозначен как параметр сонаправленности доменов.

The magnetostriction (MS) phenomenon is traditionally believed to be connected with the lattice parameter changes when a magnetic ordering is set in a crystal (spontaneous striction) or when the magnetization is changed in a magnetic field (forced striction) [1]. The forced MS is physically different depending on magnetization of a magnetically homogeneous state or a magnetically heterogeneous one with different directions of spontaneous striction in individual fragments of the substance (domains), the heterogeneity resulting from spontaneous striction at the magnetic ordering occurred in the absence of external magnetic field. These traditional concepts are to be somewhat stipulated for the case of ferromagnetic martensites exhibiting unusually high MS values [2]. The martensitic transformation, being a structure phase transition in its essence, gives rise to fragments of the martensite polycrystal structure diffing in their orientations ("variants"). Distortions thereof in relation to the initial cubic austenite structure are not caused by the spontaneous MS. After the ferromagnetic ordering, however, the magnetic anisotropy directions of the crystalline fragments turn out to be connected rigidly with the straining directions of the initial austenite structure. Thus, the anomalous large MS of the miltivariant martensite is fully equivalent to the reorientation of distortions forced by an external field, exactly as if the distortions would be caused by spontaneous MS with different directions in individual variants. In a magnetic field, the large value of magnetoelastic interactions results in a transition of the whole sample to one type of variants where the distortion direction of the initial austenite structure will be in consistency with the resulting magnetization direction, so that a giant strain of the sample will occur according to the field direction. Thus, an anomalous large MS will always take place when an external magnetic field turns the differently oriented spontaneous strictional strains of magnetic domains towards one and the same direction. This occurs independent of what is the primary phenomenon: elastic distortions (as in martensite where those arise in absence of ferromagnetic ordering) or spontaneous striction

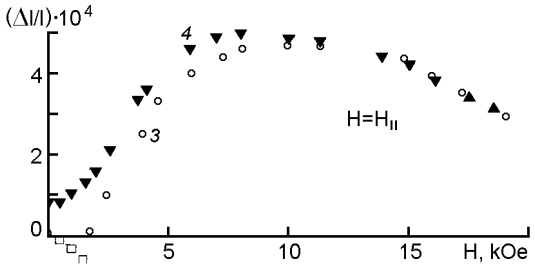

Fig. 1. MS as a function of H for $CoCl_2$ at T = 4.2 K. Curves 1, 3 answer to application of H, 2 and 4, to removal of H.

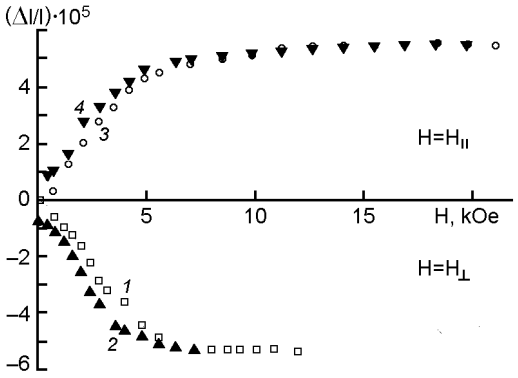

Fig. 2. MS as a function of H for $NiCl_2$ at T = 4.2 K. Curves 1, 3 answer to application of H, 2 and 4, to removal of H.

arising due to magnetic ordering. Of importance is the conservation of rigid unique relationship between main axial directions of the arising spontaneous strains and the magnetization direction. In a more general case, a similar ordering of spontaneous MS strains may occur also in antiferromagnetics. The difference is that in the latter case, a rigid relation between directions of spontaneous striction and antiferromagnetism vector. The external magnetic field should be able to adjust the antiferromagnetism vector directions in different domains. To that end, the crystal magnetic structure (spontaneous MS strains being neglected) should have spatial degeneration for different possible directions of antiferromagnetism vector, as in the case of easy-plane anisotropy. Required is also a formation mechanism of antiferromagnetic domains with different directions of spontaneous striction (and the antiferromagnetism vector, respectively) in that "easy" plane. It is just the consideration and experimental study of such a situation that is the purpose of this work.

An MS corresponding exactly to the above situation is observed in easy-plane antiferromagnetics of iron group dihalides [3]. In those crystals, the antiferromagnetic ordering is realized as a multidomain state [4]. It is clear that the magnetic interactions are insufficient to explain the origin of this state. On the contrary, the exchange energy losses when passing from one domain to other having a different orientation of the antiferromagnetism vector (**L**) require more likely a homogeneous antiferromagnetic state when the antiferromagnetism vector is oriented uniformly in the whole crystal volume. In those crystals, however, the magnetoelastic interactions are very strong and a considerable spontaneous anisotropic MS is observed therein. In multidomain state, the striction directions in the domains are different, so the crystal as a whole remains unstrained. In a magnetic field, under transition to a homogeneous state, the spontaneous striction is recovered for the crystal as a whole, so it becomes strained correspondingly.

It is to suppose that the multidomain state of easy-plane antiferromagnetics that, in contrast to the multidomain ferromagnetics, is not due to magnetostatic effects, is of magnetoelastic nature. Such a multidomain state was referred to as magnetoelastic multidomain state of antiferromagnetics.

In Figs. 1 and 2, presented are the forced MS dependences for $CoCl_2$ and $NiCl_2$ crystals obtained by measuring relative elongation in the crystal basal plane at T = 4.2 K under application/removal of magnetic field in the "crossed fields" configuration. The H field is directed along the striction measurement direction and it is just the curves with positive ε values that answer to that field, while $H_\perp$ lies also in the easy plane but is perpendicular to the measurement direction. The curves with $\varepsilon < 0$ answer to that field. It is seen that the forced MS of the crystal in the multidomain state is anisotropic and changes its sign as the field direction is changed to the transversal one. In fields exceeding 10 kOe, the transition into homogeneous state occurs. If the field absence, that is, at $H = 0$, the multidomain state with anisotropic striction compensated for the crystal as a whole takes place. Therefore, the crystal as a whole is essentially unstrained. The hysteresis of the $\varepsilon(H)$ dependence evidences the presence of an irreversible striction component. This is associated with a some excess domains where the vectors **L** remain perpendicular to the field applied before when the field is removed.

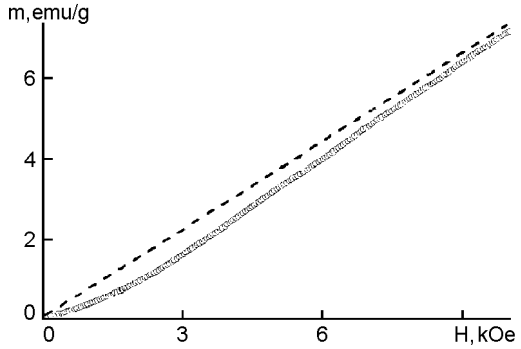

Fig. 3. $m(H)$ dependence for $NiCl_2$ at $T = 4.2$ K.

For small H, in the very onset of the rearrangement process of the multidomain state ($H \to 0$), ε depends on H quadratically:

$$\varepsilon = \varepsilon_r + \varepsilon_s \frac{H^2}{H_d^2}, \qquad (1)$$

where ε_r is the residual striction after the field removal; ε_s, spontaneous striction of single-domain state; H_d, an empirical parameter. For $CoCl_2$, $H_d^2 \approx 9.5$ kOe^2 while for $NiCl_2$, $H_d^2 \approx 13$ kOe^2.

Fig. 3 shows the magnetization dependence $m(H)$ for $NiCl_2$ at $T = 4.2$ K. According to the Neel theory [5], if an easy-plane antiferromagnetic crystal would have a homogeneous state with $\mathbf{L} \perp \mathbf{H}$, then the magnetization in a field perpendicular to the hard axis should increase linearly (dashed line) with field increasing. It is seen that the multidomain state rearrangement results in a nonlinear $m(H)$ dependence, the magnetic susceptibility in multidomain state at $H \to 0$ being almost a half of that for homogeneous one. At the same time, no irreversibility effects resulting in a loop in $\varepsilon(H)$ are observed at the magnetization measurements. At low H, $m(H)$ is described as

$$m = \chi_d H \left(1 + \frac{H^2}{H_m^2}\right), \qquad (2)$$

where χ_d is the magnetic susceptibility of multidomain state at $H \to 0$; H_m, an empirical parameter, experimental value thereof is equal to H_d.

Since both magnetization and striction are manifestations of one and the same process of the multidomain state rearrangement, the $\varepsilon(H)$ and $m(H)$ dependences are interrelated quantitatively. To establish that interrelationship, let the domain structure be assumed planar. This assumption is not in contradiction to the layered crystal structure when metal ions within the layers are bonded by ionic-covalent bonds while the layers are connected together by weaker van-der-Waals bonds. Let the domain density distribution be determined from the relation $p(\varphi) = dV_\varphi / V d\varphi$ where φ is angle between the domain magnetization direction and the field one; dV_φ, volume of domains having that orientation determined on the $d\varphi$ interval; V, the crystal volume. Since the spontaneous striction of the domains is anisotropic (it amounts ε_s perpendicularly to $\mathbf{L}$ while has the opposite sign along $\mathbf{L}$ and is equal to $-\varepsilon_s$), the average striction of the crystal is

$$\varepsilon = \frac{1}{\pi} \int_{-\pi/2}^{\pi/2} \varepsilon(\cos^2\varphi - \sin^2\varphi) p(\varphi) d\varphi = \frac{\varepsilon_s}{\pi} \int_{-\pi/2}^{\pi/2} 2(\cos^2\varphi - \frac{1}{2}) p(\varphi) d\varphi. \qquad (3)$$

At $T = 4.2$ K, modules of $\mathbf{s}_1$ and $\mathbf{s}_2$ vectors are the same: $|\mathbf{s}_1| = |\mathbf{s}_2| = s$. Then, the crystal magnetization is

$$m = \frac{1}{\pi} \int_{-\pi/2}^{\pi/2} \chi_e H \cos^2\varphi p(\varphi) d\varphi, \qquad (4)$$

where χ_e is the magnetic susceptibility of homogeneous state.

The expressions (3) and (4) make it possible to obtain a relationship between the average magnetization and striction values:

$$m = \frac{1}{2} \chi_e H \left(1 + \frac{\varepsilon}{\varepsilon_s}\right). \qquad (5)$$

Using (5), we can obtain the $m(H)$ dependence as expected from experimental data on striction $\varepsilon(H)$ [6]. In Fig. 4, such a dependence is constructed basing on the striction data for the curve 4 in Fig. 2 (for $NiCl_2$ crystal). The m(H) dependence so obtained in nonlinear and has a "sagged" shape, thus agreeing with the actual magnetization curve of Fig. 3. It follows from (5) that at $H = 0$, $m = 0$ even if the average striction for the crystal is nonzero, $\varepsilon(H = 0) \neq 0$. Therefore, no residual magnetization is observed in $m(H)$, in contrast to $\varepsilon(H)$.

It follows from (3) and (4) that the behavior of $m(H)$ and $\varepsilon(H)$ is defined by an averaged characteristic of domain orienta-

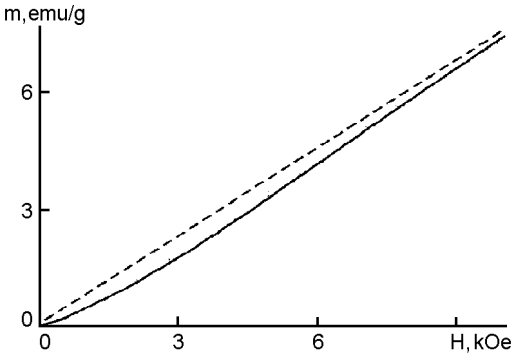

Fig. 4. $m(H)$ dependence constructed basing on $\varepsilon(H)$ data of curve 4, Fig. 2.

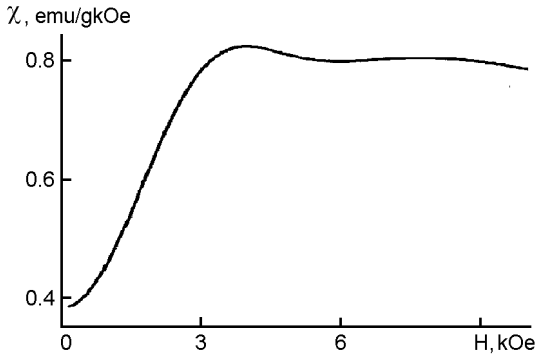

Fig. 5. Magnetic susceptibility curve for $NiCl_2$ in multidomain state.

tion that will be referred to as the domain co-alignment parameter [7] determined as

$$\bar{n}_L = \frac{1}{\pi}\int_{-\pi/2}^{\pi/2} 2\left(\cos^2\varphi - \frac{1}{2}\right) p(\varphi)d\varphi. \qquad (6)$$

The expression (6) characterizes the extent of domain co-alignment in the multidomain antiferromagnetic state. At $H = 0$, in the multidomain state $\bar{n}_L = 0$, in single-domain one, $\bar{n}_L = 1$. The average striction and magnetization are in proportion to that parameter:

$$\varepsilon = \varepsilon_s \bar{n}_L, \quad m = \frac{1}{2}\chi_e H(1 + \bar{n}_L). \qquad (7)$$

Using the introduced parameter, it is possible to describe phenomenologically the multidomain state rearrangement in magnetic field. Let the phenomenological expression for the free energy of the multidomain antiferromagnetic state be written as

$$F = a\bar{n}_L^2 + c\bar{n}_L^3 + b\bar{n}_L^4 - \frac{\chi_e}{4}H^2(\bar{n}_L + 1), \qquad (8)$$

where $a > 0$, $b > 0$, $9c^2 < 32ab$. The last item in (8) defines the interaction energy of domains with the magnetic field.

Let the simplest "quadratic" model be considered when $b = 0$, $c = 0$ in (8). This approach is identical with the model considered before in [8] where it is stated that the multidomain state is stabilized by the contribution to the free energy, which is in proportion to the squared average striction. But, as follows from (7), the squared average striction in the multidomain state is in proportion to the squared domain co-alignment parameter.

According to the quadratic model, the multidomain state is realized only on the interval $H \in [0, H_d]$. Therewith, the magnetic susceptibility increases in proportion to squared H while in the monodomenization field $H = H_d$ it becomes jump-like halved. Thus, in strong fields at the final stage of the multidomain state rearrangement, the quadratic model is inconsistent with the experimental magnetic susceptibility data (presented in Fig. 3 for $NiCl_2$ crystal).

The quadratic model, however, can be extended over the whole region of the multidomain state rearrangement fields in the case of the magnetoelastic mechanism of the multidomain state that is based on elasticity matching of domains and defects [9], if the inevitable (and even obligatory) scattering of the monodomenization field $H = H_d$ is taken into account. Under this condition, the magnetic susceptibility becomes continuous at the transition to homogeneous state (the jump in $\chi(H)$ disappears). The case $\sqrt{\sigma}/\overline{H}_d \sim 1$ (where σ is the scatter dispersion of H_d and $\overline{H}_d$ is the mean value thereof) answers to a rather good consistency with experimental $\chi(H)$ data for $NiCl_2$ crystal showing a low but very broadened maximum.

It is to note that the phenomenological model (8) in its most general form with $b \neq 0$ and $c \neq 0$ allows also to describe the multidomain state except for the field region (about 10 kOe) of transition to homogeneous state when $\bar{n}_L \to 1$. Therefore, the quadratic model taking into account the non-uniformity of monodomenization fields due to action of defects seems to be more convincing.

Thus, it has been shown that the anomalous striction behavior at the rearrangement of the multidomain state in easy-plane antiferromagnetic iron group dichlorides is associated, first of all, with arrangement of differently oriented spontaneous striction strains in domains along one and the same direction preset by the external field.

Therewith, the unique relationship between the spontaneous striction directions in domains and directions of **L** and **M** vectors therein results in a specific interrelation between the field dependences not only of the crystal average strain but of its average magnetization. The average striction and magnetization turn out to be in proportion to the domain co-alignment parameter that characterizes their average orientation. Introduction of that parameter makes it possible to describe phenomenologically the multidomain state of an antiferromagnetic in external magnetic field.

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Аномально велика магнітострикція, обумовлена перебудовою магнітопружних доменів

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Показано, що аномально великі величини магнітострикції при перебудові багатодоменного стану легкоплощинних антиферомагнетиків дигалідів групи заліза пов'язані з магнітопружною природою їх багатодоменного стану. Для опису такого багатодоменного стану введено параметр, що характеризує середню орієнтацію доменів, який позначено параметром співнаправленості доменів.