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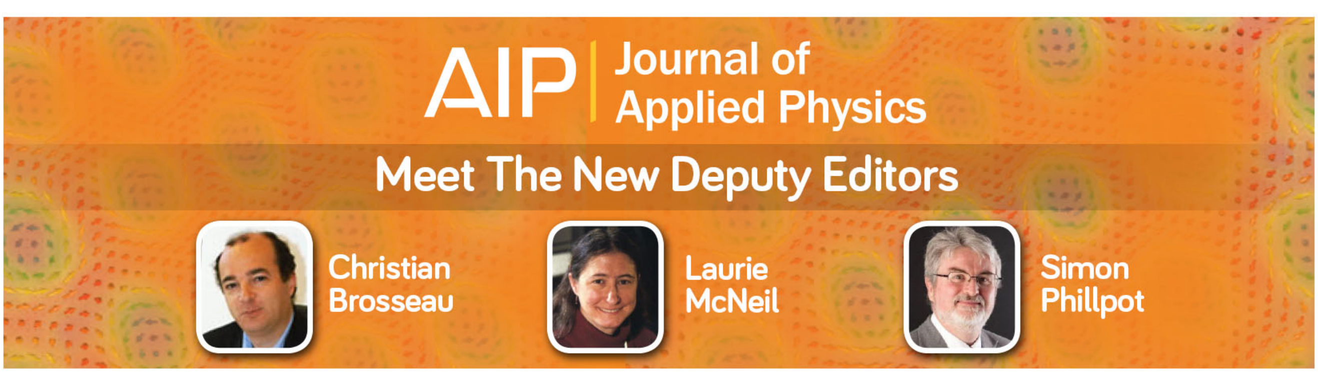

A study of the dynamics of magnetic disaccommodation in amorphous ferromagnets. I. Experimental results

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Systematic room-temperature measurements of the aftereffect of the ac magnetic permeability have been performed on a $Fe_{81}B_{13.5}Si_{3.5}C_2$ amorphous ribbon in order to get detailed information about the nature of the atomic ordering processes responsible for the magnetic relaxation. The magnetic aftereffect related to 180° domain-wall motion has been measured by means of a specific technique allowing periodic domain-wall displacements to be induced between two fixed, neighboring equilibrium positions by applying a square-wave field of proper amplitude and frequency. In this way, the average direction of the magnetization vector is cyclically modified in all points where the studied directional ordering processes may occur. As a consequence, the kinetics of ordering is modified in a characteristic way, giving rise to relevant variations in the intensity of the magnetic aftereffect measured between fixed times ($t_1 = 2\times10^{-3}$ s and $t_2 = 10^{-1}$ s), and in the value of the magnetic induction at the time t_2. All measurements have been performed at constant applied field. The degree of reliability of this experimental technique has been analyzed in detail. The magnetic aftereffect, $\Delta B = B(t_1) - B(t_2)$, and the magnetic induction $B(t_2)$ have been measured as functions of the number of domain-wall cycles, and after removing the square-wave field for a variable time t^*. The results of many independent measurements are reported and discussed.

I. INTRODUCTION

The reversible relaxation of the magnetic permeability (magnetic aftereffect or disaccommodation) is a distinctive feature of ferromagnetic metallic glasses. A number of papers dealing with this subject outline analogies and distinctive features of this relaxation with respect to the similar effect observed in some crystalline materials.[1-3] As is well known, the aftereffect of the magnetic permeability originates from a progressive damping of domain-wall motion after any domain-wall rearrangement. The damping term derives from the reorientation of mobile atom pairs or, more generally, groups of atoms, usually pictured as magnetic defects randomly distributed within the material and interacting with the local magnetization direction within each domain wall.

The nature of the coupling between magnetic defects and magnetization has been investigated in various works.[1,2,4] The microscopic ordering phenomena involved in the magnetic aftereffect have been pictured as classical two-level systems.[5] The peculiar kinetic aspects of this relaxation[3] are generally explained by involving the existence (at any temperature T) of a broad, continuous distribution of time constants τ for the directional ordering processes, weighed by a suitable distribution function $p(\tau)$. Actually, the relaxation of the magnetic permeability in amorphous ferromagnetic alloys exhibits a semilogarithmic behavior over rather extended time intervals (up to three orders of magnitude).[6] As a matter of fact, relaxation processes characterized by a semilogarithmic kinetics are very common in amorphous systems, and have been extensively studied.[7]

In the case of the magnetic aftereffect, it is possible to deal with a kinetics substantially reversible at room temperature, and covering a very wide time interval: from about 10^{-4} s to about 10^6 s after an impulsive domain-wall rearrangement. These features are suitable for checking the assumptions of the current models for the aftereffect, essentially based on the assumption that the ordering processes, characterized by a wide range of activation energies, are independent.[3,8] Such a point of view is rather common to various approaches to relaxation effects in amorphous metals.[9]

The occurrence of a magnetic relaxation indicates that a directional ordering of magnetic defects is taking place after a domain-wall rearrangement. As a consequence, aftereffect measurements give direct information on the ordering kinetics for a system of independent magnetic defects, evolving towards an equilibrium state determined by the direction assumed by the magnetization vector at a given time. Such a process may be viewed as the evolution of a thermodynamic system from an initial, off-equilibrium situation towards equilibrium. At a given time ($t = 0$), a single external parameter (the local magnetization direction) is instantaneously varied, and the system starts relaxing towards the lowest free-energy state compatible with the new value assumed by the external parameter. The slow kinetics characterizing the diffusive ordering process enables one to easily perturb the relaxation of the system of magnetic defects towards the equilibrium state.

The purpose of the present work is to perform a detailed kinetic analysis of the magnetic permeability aftereffect in a typical Fe-based amorphous ferromagnetic alloy. Part I contains the description of an experimental method, allowing one to modify the conventional ordering process by means of a controlled variation of the external parameter (the local magnetization direction), defining at every time the equilibrium state of the system of ordering defects. The specific experimental conditions required to perform these kind of measurements are discussed in detail. Typical results concerning cyclic perturbations of the ordering process for defects are reported and analyzed.

In part II, the theoretical predictions of the conventional model will be examined in detail, and shown to be able to explain the essential characters of this kinetic aspect of the magnetic aftereffect in amorphous systems.

II. EXPERIMENT

Let us first outline the adopted experimental procedure. A sample of amorphous ferromagnetic ribbon is kept saturated for a time longer than the total duration of the subsequent measurement (15 min in our case). The magnetization may be considered as essentially uniform in the saturated material. This condition (saturated sample) will be regarded as a reference state in all subsequent considerations. After removal of the saturating field at $t = 0$, a number of planar 180° domain walls nucleate in the amorphous material and are forced to oscillate around their equilibrium positions by a high-frequency ($\nu_H = 2\times10^5$ s^{-1}) driving field H_e of low amplitude ($H_e < 1$ A/m). The oscillation amplitude is of the order of one quarter of the domain-wall width, corresponding, as known, to the maximum value of the magnetic aftereffect.[1] Within any domain wall, the magnetization direction is changed from its reference-state value at $t = 0$.

During the whole measurement, the samples were submitted to a strong tensile stress along the ribbon axis in order to obtain a simple domain structure, composed of a few domains of antiparallel magnetization. The actual magnetic structure of the stressed samples was checked through a conventional Kerr-effect apparatus.

The domain walls may be periodically removed from their equilibrium positions for a given time and brought back to the original position. As a result, the magnetization alternately takes two directions, one of which is coincident with the reference-state direction (see Fig. 1).

The domain walls are cyclically displaced at low frequency ($\nu_Q = 1$ s^{-1}) between two fixed, contiguous positions [(a) and (b) in Fig. 1] by applying a suitable square-wave field Q. The permanence time of each wall in either position is $t_0 = 0.5$ s. The experimental details of the required setup have been discussed in a previous paper.[10] The amplitude of the periodic domain-wall displacement, obtained from direct measurements of B when the number of moving walls is known, turned out to be of a few micrometers, i.e., much greater than the domain-wall thickness (about 0.1 μm for this material under stress). The measurements have been performed on different as-cast strips (length 10 cm, width 0.5 cm, thickness 50 μm) of the amorphous alloy $Fe_{81}B_{13.5}Si_{3.5}$ C_2 (saturation magnetostriction $\lambda_s = 35.6\times10^{-6}$ at room temperature).

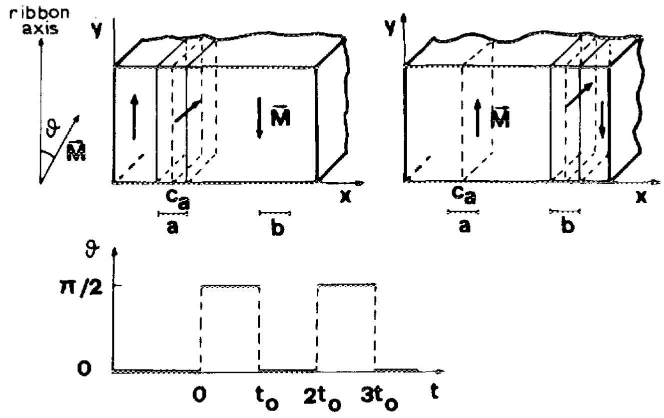

FIG. 1. Schematic picture of a domain wall oscillating between two fixed positions (a) and (b) in a ferromagnetic amorphous sample; θ is the generic angle between the magnetization and the ribbon axis. $t_0 = 0.5$ s is the half-period of the square-wave field. The direction of M in the plane $x = c_a$, corresponding to the center of the wall displaced in position (a), is sketched for both wall positions.

Subsequent decays of the magnetic induction B have been measured as functions of the number of domain-wall cycles of the type (a)-(b)-(a) after removal of the saturating field. The detecting setup was triggered in order to record only the decays corresponding to domain-wall oscillations around one equilibrium position, say, position (a), irrespectively of the number of performed cycles. A schematic picture of the observed decays is shown in Fig. 2. Note that only the decays corresponding to an even number ($2N$) of square-wave field half-periods (full lines in Fig. 2) are actually recorded. The magnetic relaxation was followed between the fixed times $t_1 = 2\times10^{-3}$ s and $t_2 = 0.1$ s, starting from every domain-wall displacement from position (b) to position (a). The driving-field amplitude, H_e, was kept fixed to the value corresponding to the maximum of the ΔB-vs-H_e curve, where $\Delta B = B(t_1) - B(t_2)$.[1]

In order to obtain coherent results, all domain walls in

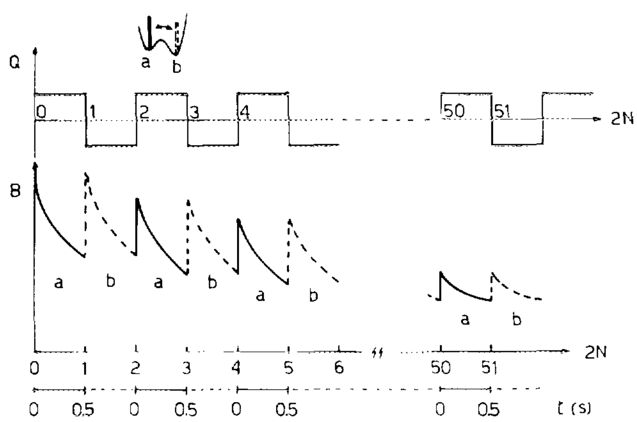

FIG. 2. Schematic picture of the square-wave cycles (above) and of the related set of measured decays (below, full lines). The decays corresponding to the permanence of the walls in position (b) are also shown (dashed lines).

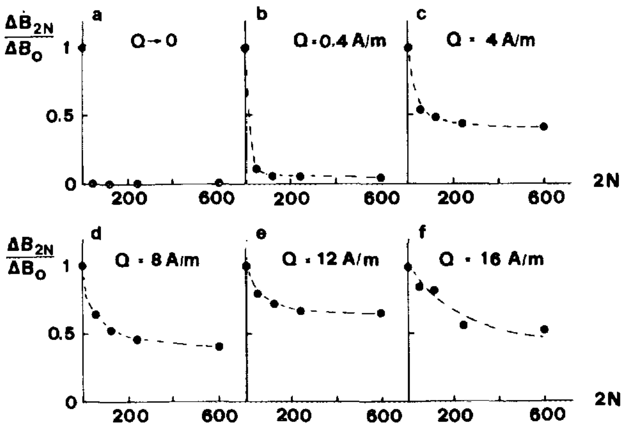

FIG. 3. Normalized magnetic relaxation, $\Delta B_{2N}/\Delta B_0$, as a function of the number of half-periods of the square-wave field, $2N$, for different values of the square-wave field amplitude Q.

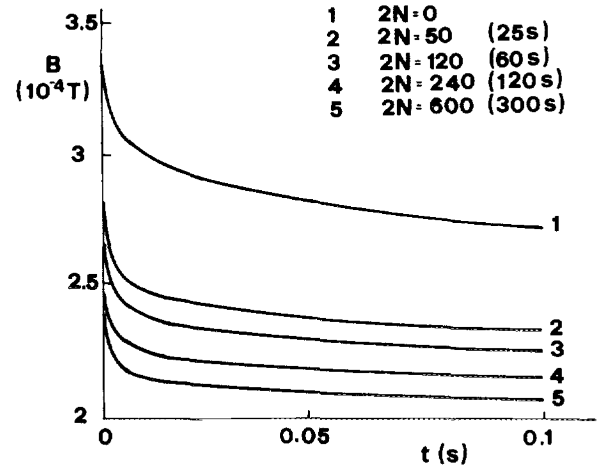

FIG. 4. Time behavior of the magnetic induction, $B(t)$, measured in the interval 2×10^{-4} s $< t < 10^{-1}$ s for various values of $2N$.

the sample must be really cycling between two fixed positions. If the positions (a)-(b) of the walls would be continuously changing with $2N$, each subsequent decay of B would be identical to the first one, since each time the walls would experience the same viscosity field, unaffected by any previously occurring diffusion process. This condition defines a lower and an upper limit of the range of suitable values for the intensity of the square-wave field.

Square-wave fields smaller than the lower limit are not sufficient to overcome the binding energy of the walls to their local pinning centers, while fields sensibly greater than coercivity give rise not only to domain-wall displacements with random character, but also to dissipative effects related to the presence of a significant eddy-current field,[11] superimposed to the conventional diffusive-type aftereffect.[12]

By considering a set of measurements of the magnetic relaxation $\Delta B_{2N} = B_{2N}(t_1) - B_{2N}(t_2)$ as functions of both the number $2N$ of square-wave field half-periods and of its intensity Q, it may be concluded that the condition of a regular, periodical motion of domain walls is verified over a sufficiently wide range of Q values.

The value of $\Delta B_{2N}/\Delta B_0$ in the absence of the square-wave field ($Q\to0$) is reported in Fig. 3(a) as a function of $2N$. The intensity of the decay is observed to rapidly vanish with increasing $2N$, because each wall never leaves position (a), in spite of the presence of H_e. On the contrary, for $Q > 12$ A/m [Figs. 3(e) and 3(f)], the dependence of ΔB on $2N$ becomes less definite. This result may be interpreted in terms of partially stochastic domain-wall cycling: in fact, when the domain walls are displaced at random by the square-wave field (e.g., $a_1 - b_1 - a_2 - b_2 - \cdots - a_n - b_n -$), the measured relaxation of the magnetic permeability is substantially independent of $2N$, since any decay is in practice the first one for any new position a_i.

It is then possible to conclude that the most convenient range of Q values to deal with is between about 1.2 and 10 A/m. There, every domain wall may be realistically assumed to perform periodic displacements between the same positions, (a) and (b), during the whole measurement. The present measurements were therefore performed at an intermediate square-wave field value, $Q = 4$ A/m.

Finally, it should be remarked that this type of measurement exhibits an intrinsic stochastic character, owing to the lack of control on domain-wall nucleation at $t = 0$. As a matter of fact, the average mobility and number of domain walls may differ after each saturation, giving rise to an unavoidable scattering of the experimental data, which may be accounted for only by performing a statistical analysis of the results.

III. RESULTS

The magnetic relaxation was measured, respectively, at the times $t = 0^+$, 5, 10, 25, 60, 120, and 300 s, after nucleation of new domain walls, i.e., after $2N = 0$, 10, 20, 50, 120, 240, and 600 reversible domain-wall displacements between the contiguous positions (a) and (b). A set of selected decays of the magnetic induction, detected according to the procedure outlined in the previous section, is shown in Fig. 4. B was monitored continuously between $t_i = 2\times10^{-4}$ s and $t_2 = 10^{-1}$ s. The data in the interval $t_i < t < t_1 = 2\times10^{-3}$ s were often affected by spurious disturbances and were consequently not analyzed, although they are shown in Fig. 4.

The same curves are reported in Fig. 5 as functions of the logarithm of time in the interval 10^{-3} s $< t < 10^{-1}$ s. Such a logarithmic plot clearly shows that no relevant changes in the shape of the relaxation curves may be observed. Only the intensity of the magnetic relaxation appears to be significantly reduced with increasing $2N$. Note that the values of $B_{2N}(t_1)$ and $B_{2N}(t_2)$ steadily decrease as a function of $2N$. Such a result may be easily understood by taking into account that the domain walls are actually cycling between the same positions, where the viscous damping is progressively increasing with $2N$, as schematically shown in Fig. 2. If instead the walls were cycling between continuously changing positions, all the measured curves should be identical to the first one. Note finally that the B

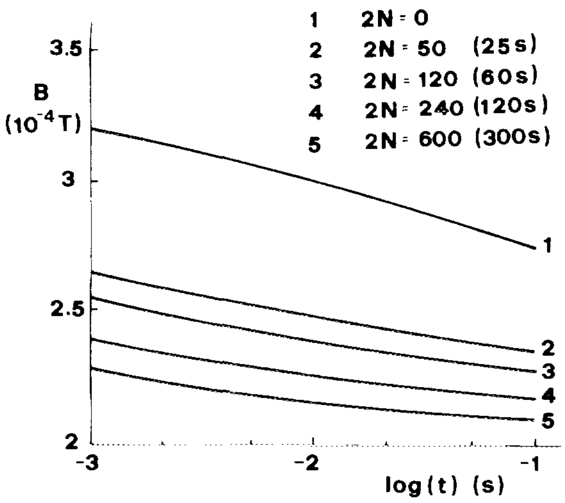

FIG. 5. Same curves as in Fig. 3, plotted vs the logarithm of time in the interval $-3 < \log t < -1$.

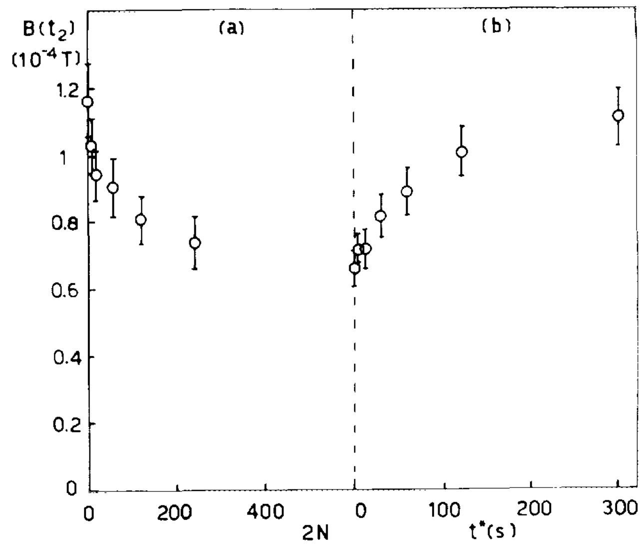

FIG. 7. Same as in Fig. 5 for the magnetic induction at $t_2 = 10^{-1}$ s.

values of the curves reported in Figs. 4 and 5 are affected by a relevant air flux contribution.

The kinetics of this relaxation may be studied by simply measuring the difference $\Delta B_{2N} = B_{2N}(t_1) - B_{2N}(t_2)$ as a function of the number of domain-wall cycles. ΔB_{2N} is observed to rapidly decrease after the first run ($2N = 0$), reaching a quasi-saturation value for times longer than about 120 s [see Fig. 6(a)]. On the other hand, the magnetic induction measured at the time t_2 is also observed to be rapidly reduced with increasing the number of domain-wall cycles [see Fig. 7(a)]. In fact, these aftereffect measurements are performed at a fixed value of the high-frequency driving field H_e. The growing hindrance to domain-wall motion, pictured in terms of a "viscosity field,"[13] reduces progressively the oscillation amplitude of domain walls (proportional to the magnetic induction B). The values of ΔB_{2N} and $B_{2N}(t_2)$ reported in Figs. 6 and 7 were properly corrected for the air flux.

A complementary measurement was performed in order to check the reversibility of the directional ordering process. The walls were left in position (b) for a given time t^*, after a fixed number of (a)-(b)-(a) cycles ($2N = 600$), by interrupting the square-wave field. The B signal was then measured with the walls again in position (a), after the time $t_{tot} = 2(N+1)t_0 + t^*$ since the beginning of the measurement. The average values of ΔB_{600} and $B_{600}(t_2)$ are shown in Figs. 6(b) and 7(b) as functions of t^* ($t^* = 3$, 10, 30, 60, 120, and 300 s, respectively).

It should be explicitly noted that the data obtained from repeated measurements under the same conditions appear to be quite scattered, owing to the mentioned stochastic character of domain-wall nucleation. As a consequence, only average values of the measured quantities will be considered in the following. The set of results reported in Figs. 6 and 7 was obtained from three different samples cut from the same amorphous ribbon in order to ensure a satisfactory degree of reliability to our measurements.

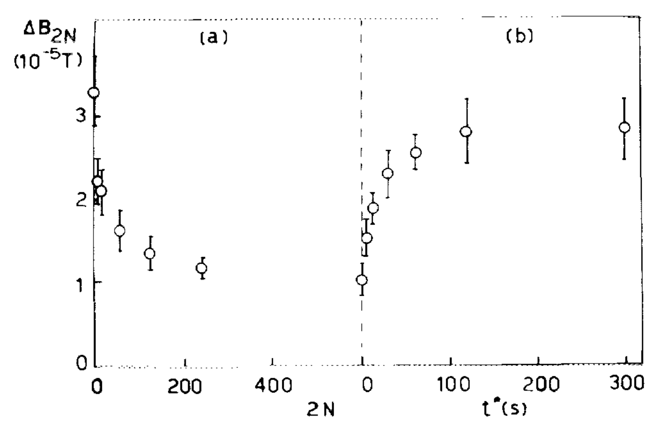

FIG. 6. Average values and standard deviations of the quantity $\Delta B_{2N} = [B(t_1) - B(t_2)]$ as functions of $2N$ (a) and of the waiting time t^* (b).

IV. CONCLUDING REMARKS

The present results indicate that the decay of the magnetic induction B at constant magnetic field between fixed times, $\Delta B_{2N} = B_{2N}(t_1) - B_{2N}(t_1)$, may be of the same order of magnitude as $B_{2N}(t_2)$ [or $B_{2N}(t_1)$], quite independently of the value of the parameter $2N$. This is a distinctive feature of magnetostrictive metallic glasses, contrasting with the behavior observed in crystalline alloys, where usually $\Delta B \ll B(t_2)$. As a matter of fact, the amplitude of the total relaxation of B_0, $\Delta B_0 = B_0(0) - B_0(\infty)$, is larger than $B_0(\infty)$, as it can be deduced from a measurement of the whole $B_0(t)$ curve for $Fe_{81}B_{13.5}Si_{3.5}C_2$, performed between 10^{-4} and 6×10^5 s, as reported elsewhere.[6] The extrapolated values of $B_0(0)$ and $B_0(\infty)$ turn out to be $B_0(0) = 1.59\times10^{-4}$ T, $B_0(\infty) = 2.44\times10^{-5}$ T for the considered samples. The ratio $\Delta B_0^{tot}/B_0(\infty)$ is as large as 5.5 in this case.

In the presence of such an intense relaxation effect, the kinetics of the progressive ordering of magnetic defects cannot be related in a simple way to the time behavior of $B_0(t)$, as shown elsewhere.[14] Information about the ordering process may however be obtained from a single measurement of $B_0(t)$ by means of an ad-hoc treatment of the experimental data.[14]

It may also be concluded that the main effect of domain-wall cycling consists of a reduction in the slope of the $B_{2N}(t)$ curve between t_1 and t_2 without a relevant change in its shape. The reduction rate steadily decreases with increasing $2N$. A similar trend is also observed for the magnetic induction measured at a given time, $B_{2N}(t_2)$.

The typical behavior of both $B_{2N}(t_1)$ and $B_{2N}(t_2)$ will be compared in part II with the predictions of a model, where the ordering magnetic defects are treated as noninteracting. It will be shown that such a model is in satisfactory agreement with our results.

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