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A comprehensive approach to broadband characterization of soft ferrites.

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Abstract

We present and discuss methods, setups, and results concerning the characterization of Mn-Zn and Ni-Zn ferrites in the frequency range DC - 1 GHz, by which we bring to light the physical mechanisms responsible for the observed frequency behavior of magnetic losses W and permeability μ and provide thorough assessment of the broadband response of the material. A comprehensive array of polarization J_p and frequency f values is investigated. A fluxmetric approach is applied up to a few MHz, giving way to a transmission line method at higher frequencies, up to 1 GHz. The fluxmetric measurements are made at defined J_p value, typically from a few mT to some hundred mT. The waveguide characterization, centered on the use of a network analyzer, is instead made under defined exciting power. But a full experimental $W(J_p, f)$ matrix up to 1 GHz and J_p values typically belonging to the Rayleigh region is in any case retrieved, thanks to the linear response of the material at high-frequencies. Disaccommodation measurements are the route followed in these experiments to separate the rotations from the domain wall process at all frequencies. Whatever the magnetization mode, the role of eddy currents in Mn-Zn ferrite losses is put in evidence by means of resistivity measurements and ensuing multiscale numerical modeling, the loss experiments being made on progressively thinned ring samples. It is concluded that an eddy current free $W(J_p, f)$ behavior can always be obtained, which can be decomposed into domain wall and rotation related contributions. The latter can be calculated assuming a suitable distribution of the effective internal anisotropy fields and its introduction in the Landau-Lifshitz-Gilbert derivation of the rotational susceptibility.

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32 1. Introduction

33 Mn-Zn and Ni-Zn sintered ferrites are the materials of choice for applications where high permeability and low 34 losses are required in a vast range of frequencies [1, 2]. But the physical interpretation of the ferrite behaviour, 35 namely magnetic loss and permeability, across the broad frequency domain useful for the various applications 36 in electrical, electronic, and telecommunication devices, lags behind today's technological developments. This 37 derives in part from lack of comprehensive magnetic characterization, typically restricted in the literature to 38 relatively narrow ranges of frequency and polarization values [3-5]. These materials do actually display a 39 complex magnetic phenomenology, where domain wall (dw) processes and magnetization rotations, combining 40 in comparable proportions under DC excitation, evolve with the magnetizing frequency through different 41 dissipation channels, descending from eddy current and spin damping phenomena. Experiments should then 42 possibly aim at identifying and separating the different types of magnetization processes and the underlying 43 dissipation mechanisms. In this paper we discuss an ensemble of methods and experiments we have developed 44 for the characterization of the magnetic properties of Mn-Zn and Ni-Zn ferrites from quasi static excitation to 45 microwave frequencies, that is, up to full relaxation of the material response. We show how fluxmetric and 46 transmission line measuring methods seamlessly combine to provide a broad matrix of values of the complex 47 permeability and the energy loss as a function of peak polarization J_p and frequency f. The role of eddy 48 currents in Mn-Zn ferrites is brought to light measuring the electrical resistivity versus f and making loss 49 measurements as a function of the sample thickness. The electrical and magnetic results are reconciled by 50 multiscale modelling of eddy current circulation in the real heterogeneous material. The ubiquitous presence of 51 the magnetic aftereffect is exploited to separate the dw and rotational contributions to the magnetization 52 process. By disaccommodation experiments performed versus frequency it is verified, in particular, that the dw 53 processes subside, in general, on entering the MHz range, and magnetization rotations prevail. These display 54 damped resonant response, with local resonance frequencies distributed according to the distribution of the 55 local effective anisotropy fields, the combination of magnetocrystalline anisotropy and internal demagnetizing 56 fields.

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Measurement of the electrical resistivity.

59 Knowledge of the electrical resistivity and its dependence on frequency is basic to the analysis of the energy 60 losses of the Mn-Zn sintered ferrites. The typically measured resistivity values are in fact compatible, contrary 61 to the case of the nearly insulating Ni-Zn ferrites, with the existence of eddy current generated losses [1, 5, 6, 62 7]. Efforts are therefore spent towards loss reduction by increasing the resistivity through addition of 63 segregating oxides, like CaO, SiO₂, Nb₂O₅, TiO₂, and fine control of the partial pressure of oxygen during 64 sintering [4, 8, 9]. It can be shown, however, that the eddy current loss contribution can be suppressed, at least 65 below a few MHz, if the sample is sufficiently mall [10]. This finding can be quantitatively assessed by 66 modelling the eddy current behaviour versus frequency, where the real heterogeneous structure of the sintered 67 material, where the semiconducting grains are separated by nearly insulating boundary layers, is taken into 68 account [11]. The capacitive properties of the nanometer-thick grain boundaries, resulting into an extremely 69 high value of the relative permittivity of the sintered material (a few 10⁴), leads to eddy currents evolving with 70 increasing frequency from local, grain boundary limited, to intergrain patterns, eventually investing the whole 71 sample cross-section [9].

72 The real ρ' and imaginary ρ'' resistivity components of the investigated Mn-Zn ferrites have been determined 73 by means of standard four-wire measurements of the electrical impedance of cut rings, where good voltmetric 74 and amperometric electrical contacts are ensured at the sample ends by sandwiching indium strips between the 75 freshly polished sample surface and the copper leads and applying uniform pressure. The temperature of the 76 sample, which was kept in a temperature controlled plastic chamber, was controlled up to T = 140 °C by means 77 of microthermocouple stuck on it. The impedance was measured up to f = 15 MHz either by an Agilent 4294A 78 LCR meter or a built-on-purpose setup employing an Agilent 33220A function generator and a 500 MHz 79 Tektronix TDS814 oscilloscope.

The simplest way to describe the electrical behavior of the Mn-Zn sintered ferrites consists in adopting the equivalent RC circuit shown in Fig. 1a, where the response of the grains and their boundary layers is described through the resistance R_g (the grains) and the parallel of a capacitance C_b and a resistance R_b (the boundary). More complex equivalent circuits can actually be envisaged, by additionally considering the capacitance of the grains and possible self-inductance effects [5] and by distinguishing between intrinsic and extrinsic grain boundaries and the related resistance-capacitance equivalent circuits [9]. It appears, however, that the circuit in Fig. 1a of impedance

$$Z(f) = R_{\rm g} + R_{\rm b} / (1 + \omega^2 R_{\rm b}^2 C_{\rm b}^2) - j \omega R_{\rm b}^2 C_{\rm b} / (1 + \omega^2 R_{\rm b}^2 C_{\rm b}^2)$$
(1)

88 can satisfactorily emulate the electrical response of all the investigated Mn-Zn ferrites, as shown by the 89 example provided in Fig. 1, where the correspondingly predicted behaviors of $\rho'(f)$ and $\rho''(f)$ for an N87-type 90 ferrite at room temperature and at T = 100 °C (dashed lines) are compared with the experiments. By 91 assimilating the sample as an ordered assembly of cubic grains of side equal to the measured average grain size 92 $\langle s \rangle$, we can estimate, according to the predicted resistivities $\rho'(f)$ and $\rho''(f)$, the resistivity of the grain ρ_g and 93 the parameters of the boundary layer: resistivity ρ_b , thickness δ , and permittivity $\varepsilon = \varepsilon_0 \varepsilon_r$. These quantities are 94 shown for the N87-type ferrite, where $\langle s \rangle = 16 \ \mu m$, in Table I. The estimated δ value, of the order of the 95 elementary cell size, appears somewhat lower than the actual grain boundary thickness, as revealed by electron 96 transmission microscopy [4, 5]. One might actually consider δ as an electrically equivalent thickness, which is 97 expected to attain the nanometer value in ferrites with high relative permittivity (say around 10^5) [12]. It is 98 observed in Table I that, according to the observed behaviour of $\rho'(f)$ and $\rho''(f)$ and Eq. (1), the increase of 99 temperature affects to considerable extent only the resistivity of the boundary layer.

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103 2. Domain wall displacements versus rotational processes.

104 Soft ferrites are characterized by very low magnetocrystalline anisotropy, resulting from the mechanism of 105 anisotropy compensation [13], and high permeability values can be obtained upon the temperature range useful 106 for applications. Magnetization rotations and dw displacements require then comparable driving fieldstrengths 107 and combine all along the magnetization curve. It is widely held, that, contrary to the case of conventional Fe-108 based polycrystalline soft magnets [14], the rotational processes may play a dominant role at low inductions 109 [15, 16], where most broadband applications lie, but it is difficult to quantify their contribution and the related 110 frequency dependence. At such inductions one may indeed take advantage of the pseudoellipsoidal shape of the 111 DC hysteresis loop and provide meaning to its evolution with frequency through the behaviour of the real u' and imaginary μ ' permeability components. The energy loss) can in such a case be expressed as 112

113 $W(J_{\rm p}) = \pi J_{\rm p}^2 \mu'' / (\mu'^2 + \mu''^2) \ [\rm J/m^3]. \tag{2}$

114 Nakamura, et al. [17, 18] proposed to separate dw μ_{dw} and rotational μ_{rot} initial permeabilities and their frequency dependence in Ni-Zn sintered ferrites by postulating resonant and relaxation response for μ_{dw} and 115 116 μ_{rot} , respectively, and making best fitting of the measured μ' and μ'' by means of five adjustable parameters. 117 The so obtained $\mu'_{dw}(f)$, $\mu''_{dw}(f)$, $\mu'_{rot}(f)$, and $\mu''_{rot}(f)$ behaviours are, however, highly hypothetical and basically 118 unproved. In this work we have separately identified the complex permeabilities $\mu_{dw}(f)$ and $\mu_{rot}(f)$ combining 119 the measurement of the initial magnetization curve with experiments on the time decay of the initial 120 permeability (disaccommodation). Examples of quasi-static initial magnetization curves, limited to their low-121 field portion ($J_p \le 50 - 100$ mT) are provided in Fig. 2a. It is the induction region of actual interest for most 122 broadband applications of soft ferrites, where, as shown in Fig. 2a, the constitutive equation J(H) closely 123 follows the Rayleigh law $J = aH + bH^2$ and the permeability is a linear function of the field $\mu(H) = a + bH$ (Fig. 124 2b). A reversible term aH, where $a = \lim_{H \to 0} \mu(H)$ is the initial permeability, and an irreversible contribution 125 bH^2 are thus identified. The irreversible magnetization is exclusively provided by the dw displacements, while 126 both rotations and dw motion combine to provide the reversible term. Assuming a uniform distribution of the 127 easy axes of the polycrystalline material, it is obtained that the rotational permeability is to good approximation 128 constant in a large part of the Rayleigh region.

129 Permeability and energy losses have been measured in Mn-Zn and Ni-Zn ring samples at different J_p values, 130 starting from $J_p = 1$ mT, from a few hundred Hz (quasi-static regime) up to about 10 MHz by means of a 131 calibrated hysteresisgraph/wattmeter. These measurements remarkably show that within the previous restricted 132 $J_{\rm p}$ limits the $\mu'(f)$ and $\mu''(f)$ curves (and, *a fortiori*, the W(f) curves) all collapse onto a single curve [19], as 133 illustrated for two J_p values in the example given in Fig. 3. This permits us to extend the experimental 134 frequency band towards the GHz range by relaxing the requirement of a defined measuring value of J_p . We 135 have applied for this a transmission line method, using a Vector Network Analyzer (Agilent 8753A), where the 136 complex permeability is obtained between about 1 MHz and 1 GHz by measuring the impedance of the ferrite 137 ring sample, placed at the bottom of a shorted coaxial line, in reflection mode under constant incident power P 138 = 10 mW [20]. Fig. 3 provides some important piece of information. Based on our previous analysis of the 139 Rayleigh diagram, we can state first that any difference $\mu'_{diff} = \mu'(J_{p2}) - \mu'(J_{p1})$, with $J_{p2} > J_{p1}$, is entirely associated with the dw processes and can be written as $\mu'_{diff} = \mu'_{dw}(J_{p2}) - \mu'_{dw}(J_{p1})$. From the observed 140 141 frequency dependence of $\mu'_{diff}(f)$ it is recognized that the dw dynamics is of relaxation type. The hump on the 142 $\mu'(f)$ curve, the landmark trait of ferromagnetic resonance, thus originates in the rotational process. It tends to 143 drown into the increasing contribution by the relaxation-type dw term μ'_{dw} with increasing $J_{\rm p}$. Coalescing of 144 the different $\mu'(J_p)$ curves at high frequencies is thus simply understood as a consequence of the decrease and 145 disappearance of $\mu'_{dw}(f)$ beyond the relaxation frequency, the surviving rotational term $\mu'_{rot}(f)$ being, as 146 previously remarked, quite independent of J_p in the Rayleigh region. The relaxation character of the dw 147 dynamics is confirmed by disaccommodation experiments, where the decay of the initial permeability versus 148 time under a weak AC field is measured immediately after the demagnetization process. Disaccommodation is 149 exclusively related to the dws, whose oscillatory motion slightly shrinks with time under the stabilizing action 150 of diffusing cations. We have shown in a previous work [11] that the rate of decay of μ ' (of logarithmic type) 151 in Mn-Zn ferrites decreases with increasing the frequency of the exciting field in a way totally consistent with a 152 relaxation dispersion of the dw permeability, eventually tending to revert, according to the theory, to a slight 153 increase beyond the cutoff frequency. This equally happens in Ni-Zn ferrites, as demonstrated in Fig. 4 by the 154 time dependence of the reduced permeability $\mu'(t)/\mu'(t_0)$, with $t_0 = 2$ s, measured at field frequencies ranging 155 between 500 kHz and 2 MHz.

Having clarified the nature of the dispersion of $\mu_{r,dw}(J_p)$, we can turn to quantitative discrimination between the dw and rotational permeabilities, again exploiting disaccommodation. The idea is one of comparing the permeability decays starting from the demagnetized state and from remanence (major loop). The former condition is attained by conventional demagnetization procedure at 50 Hz, the latter by sudden release of a saturating DC field. The strength of the weak AC exciting field is such as to engender a quasi-reversible polarization swing, typically $\Delta J = \pm 2$ mT. In the limit of small oscillations the rotational susceptibility can be written, for easy axes of anisotropy constant *K* distributed according to an angular function $g(\theta)$,

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$$\chi_{\rm rot} = \int_{-\pi/2}^{\pi/2} (\mu_0 M_{\rm s}^2 / 2K) \cdot \sin^2 \theta \ g(\theta) d\theta \,, \tag{3}$$

where M_s is the saturation magnetization. Since $\sin^2\theta = \sin^2(\pi - \theta)$, all quadrants are equivalent and χ_{rot} is the same for small moment rotations occurring either at remanence $(J = J_r)$ or in the demagnetized state (J = 0). The difference $\Delta\mu$ between the small amplitude permeabilities observed in these two states is therefore entirely ascribed to the lower domain wall contribution at remanence $\Delta\mu(t) = \mu_{dw}(J = 0, t) - \mu_{dw}(J = J_r, t)$. Fig. 5 shows that the permeability, taken at f = 10 kHz, decays slightly faster in the demagnetized state. Since the derivative $D = d\mu/d(\log t)$ can be taken approximately proportional to μ_{dw} at any time t [21], we can also write

- 170 $\mu_{\rm dw}(J=0,t)/\mu_{\rm dw}(J=J_{\rm r},t) = D(J=0,t)/D(J=J_{\rm r},t), \qquad (4)$
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where the time *t* of interest is the one where the loss measurement is performed after demagnetization, typically around 10³ s. Combining at this time the previous equations and further taking into account that with f = 10kHz $\mu(t) \cong \mu'(t)$ and $\mu'(f) \cong \mu'_{DC}$, we obtain the dw contribution to the real permeability component under quasi-static excitation

$$\mu'_{\rm dw, DC} (J=0) = \frac{\Delta \mu}{1 - D(J=J_{\rm r}) / D(J=0)},$$
(5)

at the low J_p level impressed by the AC field ($J_p = 2 \text{ mT}$ for the case of Fig. 5). We estimate for such a case the 177 178 relative permeabilities $\mu'_{r,dw,DC} = 445$ versus $\mu'_{r,DC} = 2040$ (N87 ferrite) and $\mu'_{r,dw,DC} = 695$ versus $\mu'_{r,DC} = 4540$ 179 (N30 ferrite). Having thus estimated $\mu'_{dw,DC}$ ($J_p = 2 \text{ mT}$), it is an easy matter to retrieve the whole $\mu'_{dw}(f)$ 180 behaviours at the different J_p values exploiting the previously introduced experimental quantities $\mu'_{diff}(f, J_{pi}) =$ 181 $\mu'(f, J_{pi}) - \mu'(f, J_p = 2 \text{ mT}) = \mu'_{dw}(f, J_{pi}) - \mu'_{dw}(f, J_p = 2 \text{ mT})$. We can in fact estimate $\mu'_{dw}(f, J_p = 2 \text{ mT})$ 182 assuming it replicates the frequency dependence of the experimental $\mu'_{diff}(f, J_{pi} = 5 \text{ mT})$, while tending to the 183 calculated $\mu'_{dw,DC}$ ($J_p = 2 \text{ mT}$) at low frequencies. It is then immediate to calculate $\mu'_{dw}(f, J_{pi})$ at all J_{pi} values, 184 till an upper limit roughly identified by the Rayleigh law. Quite a similar procedure, passing through the 185 etermination of the experimental quantity $\mu''_{diff}(f, J_{pi}) = \mu''(f, J_{pi}) - \mu''(f, J_p = 2 \text{ mT}) = \mu''_{dw}(f, J_{pi}) - \mu''_{dw}(f, J_p = 2 \text{ mT})$ 186 2 mT), can be applied in order to retrieve the imaginary component $\mu''_{dw}(f, J_{pi})$. Now the low-frequency limit 187 is given by the condition $\mu''_{dw,DC}$ $(J_p = 2 \text{ mT}) \equiv \mu''(J_p = 2 \text{ mT})$, because the quasi-static energy loss is 188 exclusively generated by the dw processes. Fig. 6 compares the so-extracted $\mu'_{dw}(f, J_{pi})$ and $\mu''_{dw}(f, J_{pi})$ 189 behaviours with the measured permeability components $\mu'(f, J_{pi})$ and $\mu''(f, J_{pi})$, in the J_p range 2 mT – 50 mT 190 (Mn-Zn ferrite N87). The relaxation character of the dw dynamics leads to vanishing of $\mu'_{r,dw}(f)$ and $\mu''_{r,dw}(f)$ 191 on entering the MHz range, where they are completely superseded by the rotational terms. These are associated 192 with the resonant response of the material, as illustrated by the peaked behaviour $\mu'_{r,rot}(f) = \mu'_r(f) - \mu'_{r,dw}(f)$ in 193 Fig. 7.

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195 **3.** Energy losses.

We have remarked that the phenomenology of energy losses can be viewed in terms of complex magnetic permeability, as synthetically shown by Eq. (1). The previous analysis gives then us the opportunity to discriminate between the loss contributions originating from the dw and the rotational processes.

199 Writing the expression for the energy loss at a given J_p value as

$$W(J_{\rm p}) = \pi J_{\rm p}^2 (\mu''_{\rm dw} + \mu''_{\rm rot}) / (\mu'^2 + \mu''^2) = W_{\rm dw} (J_{\rm p}) + W_{\rm rot} (J_{\rm p}), \qquad [\rm J/m^3]$$
(6)

and taking the μ''_{dw} and μ''_{rot} dependence on frequency obtained through the previously discussed procedure (see Fig. 7), we arrive at the decomposition of the loss in its dw and rotational components. Fig. 8 provides an example of the so-obtained $W_{dw}(f)$ and $W_{rot}(f)$ behaviours at two different J_p values in a Mn-Zn N87-type ring sample (outside diameter 15 mm, inside diameter 10.4 mm, thickness 5.09 mm). It is noted that $W_{dw}(f)$ and $W_{rot}(f)$ comparably contribute to W(f) in a relatively narrow intermediate frequency interval only. It is also 206 observed that, according to Eq. (6), $W_{rot}(f)$ strictly follows a J_p^2 law, being μ ''rot independent of J_p , while $W_{dw}(f)$ 207 grows faster, because μ''_{dw} increases with J_p (see Fig. 6). Consequently, overlapping between the fluxmetric 208 and transmission line measurements occurs over a shrinking region on increasing J_p . In this region, a relatively 209 sharp transition of the slope of the W(f) curve occurs. It might be argued that such a transition could be related 210 in some way to the concurring drop of the electrical resistivity (see Fig.1), thereby implying an important 211 contribution to the losses by the eddy currents. This is not the case. A same W(f) behaviour is in fact observed 212 in the insulating Ni-Zn ferrites [22] and in sufficiently small Mn-Zn samples, like the ones considered in Fig. 8. 213 Eddy current losses in the sintered Mn-Zn ferrites have important practical implications, as demonstrated by 214 the constant efforts devoted to the development of materials with increased grain boundary resistivity [4], but 215 their calculation is difficult, because of the electrically heterogeneous structure of the material. Their role is in 216 any case apparent when measuring the losses in differently sized Mn-Zn samples. The general literature 217 approach to the calculation of eddy current losses is one of applying the conventional formulation for a 218 homogeneous material (classical loss $W_{\text{class}}(J_p, f)$), which, for a circular sample of diameter d is simply 219 expressed as

$$W_{\text{class}}(J_{\rm p},f) = \frac{\pi^2}{16\rho'(f)} \cdot d^2 J_{\rm p}^2 f , \qquad (7)$$

221 where $\rho'(f)$ is the real resistivity component, as obtained with the measurements reported in Fig. 1. It has been 222 shown, however, that Eq. (7) can largely overestimate the actual eddy current losses and can be reconciled with 223 a correct formulation only at very high frequencies in [11]. Because of the insulating character of the grain 224 boundary layer, eddy currents at low frequencies circulate inside the grains and the ensuing energy dissipation 225 is much lower than the one predicted using the measured $\rho'(f)$ in Eq. (7), which amounts to assume eddy 226 current patterns freely investing the sample cross-section. The general problem of calculating the eddy current 227 losses in the sintered Mn-Zn ferrites has been discussed in [11], where the electromagnetic field equations in 228 the heterogeneous material are formulated under a variational multiscale approach and numerically 229 implemented by means of finite element method, by which the evolution of the eddy current patterns from 230 grain limited to global upon increasing frequency can be described. It is obtained, in particular, that the 231 numerical results can be fitted by the closed expression

$$W_{\text{eddy}}^{(\text{cl})}(J_{\text{p}},f) = \left\{ \frac{(k_0 - k_\infty)A}{1 + \exp[m\log_{10}(f/f_0)]} + k_\infty A \right\} J_{\text{p}}^2 f , \qquad [\text{J/m}^3]$$
(8)

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where $k_0 A J_p^2$ and $k_\infty A J_p^2$ are the calculated limits of $W_{eddy}^{(cl)}(J_p, f)/f$ for $f \rightarrow 0$ (eddy currents confined in a single grain, typically below a few kHz) and $f \rightarrow \infty$ (eddy currents circulating across the whole sample crosssection, typically beyond a few MHz), respectively. Eq. (8) provides a "classical contribution" (it will actually tend to $W_{class}(J_p, f)$ at high frequencies), because the calculation does not take into account the role of the moving domain walls and the ensuing local eddy currents. The related contribution (the excess eddy current loss) has

been estimated [10, 11] and found to be always negligible with respect to $W_{eddy}^{(cl)}$, which then accounts for the 239 240 whole of the eddy current losses. The quantity A is an effective area, depending on the width-to-thickness ratio 241 w/h. It is related to the actual cross-sectional area S as A = gS, where g is geometrical constant given by $g = 1 - 0.0923 (p-1)^{1.63}$ and p = w/h if w > h and p = h/w if h > w. Each material is associated with a defined set 242 243 of parameters k_0 , k_{∞} , m, and f_0 , where the constants m and f_0 are obtained from best fitting of the curves 244 calculated with the numerical method. Eq. (8) has been applied to differently sized Mn-Zn ring samples, as 245 illustrated in the example given in Fig. 9, where m = 4.68 and $f_0 = 3.2 \cdot 10^6$. It is noted here that only in the largest 246 sample (cross-section of width w = 5.25 mm and height h = 9.72 mm) eddy currents appreciably contribute to W(f) (at least below f = 10 MHz). By subtracting the calculated $W_{eddy}^{(cl)}(f)$ from the measured loss, we obtain the 247 248 eddy current free $W_{sd}(f)$ behavior (thick solid curves in Fig. 9). The large and the small N30-type ring samples 249 have not the very same structure, resulting into an about 30% different quasi-static loss. But at high frequencies 250 all eddy current free curves merge into a single curve, as expected for a rotations dominated magnetization 251 process. This finding permits us, with little loss of accuracy, to dispense from the complications of the numerical 252 procedure. One can either adjust the value of the constants m and f_0 for best fitting of the eddy current free curve after subtraction of $W_{\text{eddy}}^{(\text{cl})}(f)$ from the measured W(f) or exploit the general empirical finding that the ratio of 253 the conventional to the actual classical eddy current losses $R(f) = W_{class}(f) / W_{eddy}^{(cl)}(f)$ is a relaxation-type 254

function. At low frequencies this ratio is a precisely known quantity R_0 and we can pose $R(f) = 1 + \frac{R_0 - 1}{1 + (f/f_1)^2}$,

where the parameter f_1 is again obtained by best fitting of the $W_{sd}(f)$ curve. In this way we are finally left to 256 257 justify the $W_{sd}(f)$ behaviour, which, in many practical samples (as in the example given in Fig. 8), is coincident 258 with the W(f) behavior. The physical mechanism lying behind the energy dissipation is the damping of the 259 precessional motion of the spin moments, taking place both inside the moving domain walls and inside the 260 domains. The energy loss in the sample of Fig. 8 all originates from spin damping and is decomposed into dw 261 and rotational contributions. The former can be lumped in the coercivity and the additional dynamic dw effects 262 and we can write $W_{dw}(f) = W_h + W_{dw,dyn}(f)$, where W_h is the hysteresis (quasi-static) loss and $W_{dw,dyn}(f)$ can be 263 expressed in a way analogous to the standard derivation of the excess loss in metallic alloys [10, 22], the starting 264 point in both cases being an identical equation for the damped motion of the dw [23]. The spin damping 265 coefficient β_{sd} appearing in this equation is related to the Landau-Lifshitz damping constant α_{LL} by the expression $\beta_{sd} = (2J_s / \mu_0 \gamma \delta) \cdot \alpha_{LL}$, where J_s is the saturation polarization, μ_0 is the magnetic constant, γ is the 266 electron gyromagnetic ratio, and δ is the dw thickness. The viscous response to the applied AC field of the spin 267 268 moments inside the domains is described by the linear Landau-Lifshitz-Gilbert equation, by which we can obtain 269 analytical formulations for μ'_{rot} and μ''_{rot} . The pretty low values of the magnetocrystalline anisotropy of the soft 270 ferrites makes the intergrain demagnetizing fields to interfere with the magnetocrystalline anisotropy fields, resulting in a distribution of the effective internal anisotropy fields $H_{k,eff}$, that is a continuous spectrum of resonance frequencies. The theoretical μ'_{rot} and μ''_{rot} behaviours are obtained assuming that $H_{k,eff}$ is distributed

273 according to a lognormal function
$$g(H_{k,eff}) = \frac{1}{\sqrt{2\pi\sigma}H_{k,eff}} \cdot \exp[-\frac{(\ln(H_{k,eff}) - h)}{2\sigma^2}]$$
, where $h = \langle \ln(H_{k,eff}) \rangle$ and σ ,

the standard deviation of $\ln(H_{k,eff})$, are adjusted for best fitting of the experimental rotational permeabilities, once further integration is made over the isotropic distribution of the easy axes in the sample plane. An example of predictive capability provided by this approach is shown in Fig. 7, regarding the Mn-Zn ferrite type N87, for which where $\alpha_{LL} = 0.04$, h = 5.3, $\sigma = 1.2$ have been used. The rotational loss can then be immediately predicted through Eq. (6), resulting in the fitting lines shown in Fig. 10 for three different J_p values.

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4. Conclusions.

281 We have presented and discussed the broadband magnetic behaviour of Mn-Zn and Ni-Zn sintered ferrites, 282 clarifying the respective roles of domain wall displacements and rotational processes in providing the material 283 response from DC to 1 GHz. With the help of permeability disaccommodation measurements, we have been 284 able to quantitatively identify and separate the real and complex permeability components μ'_{dw} , μ'_{rot} , μ''_{dw} , and 285 μ ''_{rot} and, a fortiori, dw W_{dw} and rotational W_{rot} energy losses across the whole broad range of frequencies. 286 Evidence is offered for the relaxation character of the dw dynamics, which is completely thwarted by the 287 dissipating mechanisms on entering the MHz range and fully overcome by the rotations. It has been shown that 288 eddy currents can contribute to the magnetic losses in Mn-Zn ferrites, provided the frequency is sufficiently 289 high and the samples are large, as quantitatively predicted by a specific multiscale model relying on the 290 measurement of the electrical resistivity of the material as a function of frequency. It is concluded that, for any 291 defined peak polarization value, an eddy current free $W_{sd}(f) = W_{sd,dw}(f) + W_{sd,rot}(f)$ curve, independent of sample 292 size, can be recovered for any material, where spin damping is the associated dissipation mechanism. The 293 rotational permeability $\mu'_{rot}(f)$ and $\mu''_{rot}(f)$ curves are predicted as solutions of the Landau-Lifshitz-Gilbert 294 equation, under assumed lognormal distribution of the internal effective anisotropy fields, and the associated 295 $W_{\rm sd,rot}(f)$ behaviour is correspondingly obtained, resulting in good agreement with the experimental results.

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306 **References**

- 307 [1] E.C. Snelling, *Soft Ferrites: Properties and Applications*, Butterworths, 1988.
- V.T. Zaspalis, M. Kolenbrander, R. Guenther, and P. van der Valk, Loss mechanisms in modern MnZn
 ferrites for power applications, *Proc. 9th Int. Conf. Ferrites-9*, R.F. Sohoo, Ed., San Francisco, 2004, 579 584.
- [3] H. Saotome and Y. Sakaki, Iron loss analysis of Mn-Zn ferrite cores, *IEEE Trans. Magn.*, 33 (1997) 728–
 734.
- J. Hanuszkiewicz, D. Holz, E. Eleftheriou, and V. Zaspalis, Materials processing issues influencing the
 frequency stability of the initial magnetic permeability of MnZn ferrites, *J. Appl. Phys.* 103 (2008) 103907.
- [5] V. Loyau, G.-Y. Wang, M. Lo Bue, and F. Mazaleyrat, An analysis of Mn-Zn ferrite microstructure by
 impedance spectroscopy, scanning transmission electron microscopy and energy dispersion spectrometry
 characterizations, *J. Appl. Phys.* 111 (2012) 053928.
- [6] M. LoBue, V. Loyau, and F. Mazaleyrat, Analysis of volume distribution of power loss in ferrite cores, *J. Appl. Phys.* 109 (2011) 07D308.
- E. Cardelli, I. Fiorucci, and E. Della Torre, Estimation of MnZn ferrite core losses in magnetic
 components at high frequency, *IEEE Trans. Magn.* 37 (2001) 23666.
- [8] E. Otsuki, S. Yamada, T. Otsuka, K. Shoji, and T. Sato, Microstructure and physical properties of Mn-Zn
 ferrites for high-frequency power supplies, *J. Appl. Phys.* 69 (1991) 5942-5944.
- M. Drofenik, A. Znidarsic, and I. Zajc, Highly resistive grain boundaries in doped MnZn ferrites for high
 frequency power supplies, *J. Appl. Phys.*, 82 (1997), 333-340.
- [10] F. Fiorillo, C. Beatrice, O. Bottauscio, A. Manzin, and M. Chiampi, Approach to magnetic losses and their
 frequency dependence in Mn-Zn ferrites, *Appl. Phys. Lett.*, **89** (2006) 122513.
- [11] F. Fiorillo, C. Beatrice, O. Bottauscio, E. Carmi, Eddy current losses in Mn-Zn ferrites, *IEEE Trans. Magn.* **50** (2014) 6300109.
- 330 [12] J. Smit and H.P.J. Wijn, *Ferrites*, Eindhoven: Philips Technical Library, 1959, p. 236.
- [13] H. Pascard, Basic concepts for high permeability in soft ferrites, J. Phys. IV (France) 8 (1988) 377-384.
- [14] G. Bertotti, F. Fiorillo, and M. Pasquale, Reversible and irreversibile magnetization in soft iron-based
 polycrystalline materials, *J. Appl. Phys.*, 69 (1991), 5930-5932.
- [15] M.T. Johnson and E.G. Visser, A coherent model for the complex permeability in polycrystalline ferrites,
 IEEE Trans. Magn. 26 (1990) 1987-1989.
- [16] P. J. van der Zaag, P. J. van der Valk, and M. Th. Rekveldt, A domain size effect in the magnetic hysteresis
 of NiZn-ferrites, *Appl. Phys. Lett.* 69 (1996) 2927-2929.
- [17] T. Nakamura, T. Tsutaoka, and K. Hatakeyama, Frequency dispersion of permeability in ferrite composite
 materials, *J. Magn. Magn. Mater.* 138 (1994) 319-328.
- [18] T. Tsutaoka, M. Ueshima, T. Tokunaga, T. Nakamura, and K. Hatakeyama, Frequency dispersion and
 temperature variation of complex permeability of Ni-Zn ferrite composite materials, *J. Appl. Phys.* 78 (1995), 3983-3991.
- [19] C. Beatrice, O. Bottauscio, and F. Fiorillo, Eddy current and spin damping losses in Mn-Zn ferrites, *J. Jpn. Soc. Powder Metallurgy* 61 (2014), S208-S210.
- [20] A. Magni, F. Fiorillo, E. Ferrara, A. Caprile, O. Bottauscio, and C. Beatrice, Domain wall processes,
 rotations, and high-frequency losses in thin laminations, *IEEE Trans. Magn.* 48 (2012) 3796-3799.
- 347 [21] S. Chikazumi, *Physics of Magnetism*, Wiley, 1964, p. 303.
- [22] F. Fiorillo, M. Coïsson, C. Beatrice, and M. Pasquale, Permeability and losses in ferrites from dc to the
 microwave regime, *J. Appl. Phys.* 105 (2009), 07A517.
- [23] J.F. Dillon and H.E. Earl, Jr., Domain wall motion and ferromagnetic resonance in a manganese ferrite, J.
 Appl. Phys. **30** (1959), 202-213.
- 352 353
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- 355
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