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Studies of domain wall motion: III. The field dependence of differential susceptibility at the early stage of magnetization

W G Zeng† and G G Siu‡

† 2302, Dongguan Institute of Technology, Guangdong 511700, People's Republic of China

‡ Department of Applied Science, City Polytechnic of Hong Kong, Hong Kong

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Abstract. Differential susceptibilities χ_{diff} of nickel wire, Permalloy-42 wire, 4% silicon iron sheet and bars of SAE 1015 and 1045 steels were measured when undergoing uniform magnetic field sweeping. The field dependences of χ_{diff} of wires, sheet and bars at the early stage of magnetization were studied: in general, χ_{diff} decreases with increasing sweep rate of field and the $\chi_{\text{diff}}-H$ relationship depends on sample shape. The experimental results are interpreted using a phenomenological model based on domain wall (DW) motion that is a modest generalization of the Rayleigh law at low field. This model is also compared with a previous model of magnetomechanical damping.

1. Introduction

The dynamic behaviour of ferromagnetic materials, e.g. their response to alternating stress when undergoing uniform magnetic field sweeping (Zeng *et al* 1990, 1991, hereafter referred to as I and II), arouses great interest. In I and II, low-frequency inelastic studies of soft ferromagnetic material Fe, Ni and Permalloy-42 wires under uniformly increasing magnetic fields have been described. Their internal friction (IF) divides into static IF Q_0^{-1} and dynamic IF Q_m^{-1} . Q_m^{-1} of Fe, Ni and Permalloy are similar: $Q_m^{-1} \propto (dM/dH)(\dot{H}^s/\omega^{1+s-2\alpha})$, where M , H , ω , s and α are the magnetization, magnetic field, angular frequency of the perturbing alternating force and two index parameters respectively. A four-parameter phenomenological model was proposed to fit its H dependence. This raises further questions: whether or not differential susceptibility χ_{diff} ($= dM/dH$) depends on the field sweep rate, and whether χ_{diff} has similar field dependence to the phenomenological model. The usual magnetic measurements could not answer these questions since they involve integration of the flux change rate (Cullity 1972). This work aims at making a preliminary investigation of the field and sweep rate dependences of the differential susceptibilities of some soft ferromagnetic materials in the shapes of wires, sheets or bars. The results show that the answers to these questions are positive.

Magnetization is related to domain wall (DW) motion and magnetization rotation as first shown by Williams and Shockley's (1949) 'picture frame' experiment. The DW motion is discussed with the equation of motion of the wall per unit area, in the wall position x , which is similar to that of an oscillating system. The smoothly increasing field appears in the driving force per unit area, i.e. the magnetic energy (the principal driving force) $M \cdot Ht$, so that the DW motion and, thus, the magnetization depend not only on the magnetic field but also the field sweep rate, usually implicitly. This viewpoint succeeds in interpreting

experimental results (e.g. in I and II) and our results can be explained accordingly. The four-parameter phenomenological model is shown to be a modest generalization of the Rayleigh law of magnetization of polycrystalline specimens in low fields.

2. Experimentation

The method of measurement of χ_{diff} in our experiments differs from the ballistic method for wires or rods, which uses an electronic fluxmeter in a hysteresigraph or an RC integrating circuit in a loop tracer to integrate the signal from the search coil and then record the data automatically by an X, Y recorder or an oscilloscope. Instead, we connect the search coil directly through an amplifier to the $Y(M)$ axis of an X, Y recorder. The terminals of a fixed resistor of value R in series with the magnetizing winding are connected to the $X(H)$ axis of the recorder as usual.

The specimen, after being demagnetized slowly in a 50 Hz alternating field, is placed axially in the centre of a water-cooled solenoid 670 mm in length. On the centre of the specimen a search coil is wound. The solenoid is connected to the magnetizing circuit which includes a high-power DC supply with a device for steadily varying the current. The magnetic field sweep rate is controlled by the current sweep rate. The relationship between the magnetic field H and the current I is calibrated experimentally: H (Oe) = 32.24 I (A), which is rigorously linear to about 10 Oe. The amplification of the low-drift high-sensitivity ($5 \text{ V } \mu\text{V}^{-1}$) amplifier is also calibrated before and after measurement of each magnetization curve at a definite sweep rate, as well as the calibration curve of H versus t without a sample, and the drift curve of the amplifier. The stopwatch has a minimum division of 0.01 s.

In the CGS system of units, an EMF \mathcal{E} in the search coil is induced by any change of magnetic flux following Faraday's law: $\mathcal{E} = 10^{-8} N(d\Phi/dt) = 10^{-8} NS(dB/dt)$ where N is the number of turns and S the cross sectional area of the search coil. For ferromagnetic materials at a low drive field, $4\pi M$ is essentially equal to the magnetic induction B , i.e. $dM/dt \simeq (1/4\pi) dB/dt = 10^8 \mathcal{E}/(4\pi NS)$. In uniform field sweeping, $H = \dot{H}t$ and the differential susceptibility $\chi_{\text{diff}} = (10^8/4\pi NS\dot{H})\mathcal{E}$, which is measured along the Y axis of the recorder after being amplified A times. We obtain the $\chi_{\text{diff}}-H$ curve from the recorder after normalization.

Wire specimens included industrial Ni wire (99% pure) 2.82 mm in diameter and Permalloy-42 ($\text{Ni}_{0.42}\text{Fe}_{0.58}$) 2.02 mm in diameter, all 160 mm in length, which were all forged, rolled and annealed at 860 °C for 6 h in vacuum. The sheet sample was 4% Si steel of size 0.48 mm \times 7.71 mm \times 160 mm, a replacement for the Fe wire to obtain a measureable signal. Bar specimens were made of pure Fe (C content less than 0.05%), and SAE 1015 and 1045 high-quality steel, 6.76 mm, 7.94 mm and 7.34 mm in diameter respectively and about 190 mm in length. They were annealed at 950 °C for 4 h in vacuum. All samples were polished with corundum grease and finally annealed at 860 °C for 2 h in vacuum to reduce internal stresses caused by processing as much as possible. Average sizes of crystallites were 0.01–0.05 mm for the bar samples and 0.05–0.5 mm for the wire specimens. The field sweeping range was from 0 Oe to 10 Oe and the sweep rate was low: mostly less than 80 mOe s^{-1} and only one rate 6.5 Oe s^{-1} . In the experiments several sweep rates in the order of tens of mOe s^{-1} and one of 6.5 Oe s^{-1} were used.

3. Results and discussion

The differential susceptibilities χ_{diff} of the above-mentioned six samples were measured. All $\chi_{\text{diff}}-H$ curves decrease with sweep rate \dot{H} (the induced EMF \mathcal{E} , in contrast, increases with sweep rate \dot{H}). χ_{diff} of the wire or sheet samples reaches a maximum at a field less than 1 Oe, but that of bar samples reaches a broad plateau between 2 and 6 Oe. Then all χ_{diff} diminish toward zero in a large H . Figure 1 shows typical $\chi_{\text{diff}}-H$ curves (the Si-Fe sheet sample), which resemble the $Q^{-1}-H$ curves of Fe wire in I but within a narrower range. The curve maximum (or plateau) corresponds to the steepest part of the magnetization curve in which the change in M is mainly due to DW motion in a polycrystalline sample. Once the 'knee' of the magnetization curve is reached, i.e. χ_{diff} starts to decrease, the process of magnetization rotation predominates, though this division is not precise. A narrower field range was therefore selected for studying DW motion only. We chose the field range in which χ_{diff} changes markedly, in which mainly reversible and irreversible DW motion occurs. Figures 2 and 3 show the $\chi_{\text{diff}}-H$ curves of the samples. Those of wires and sheets are similar to each other (the field range is 0–0.5 Oe). There are Barkhausen jumps when χ_{diff} reaches the maximum but these are not shown in the graph. The $\chi_{\text{diff}}-H$ curves of bars also resemble each other in the range 0–2 Oe and no Barkhausen jumps are detected owing to the averaging over the motion of a larger number of DWs. The difference in the field range of DW motion is purely caused by the size factor of wires (or sheets) and bars.

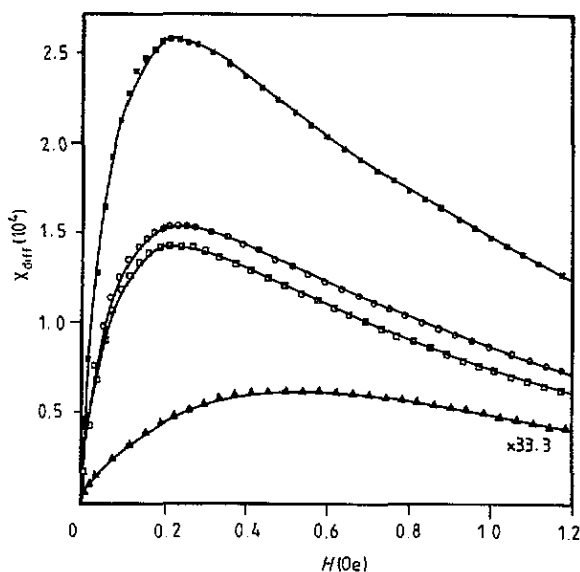


Figure 1. Susceptibility χ_{diff} of 4% Si-Fe sheet as a function of magnetic field at different sweep rates (■, $\dot{H} = 43 \text{ mOe s}^{-1}$; ○, $\dot{H} = 67 \text{ mOe s}^{-1}$; □, $\dot{H} = 75 \text{ mOe s}^{-1}$; ▲, $\dot{H} = 6.5 \text{ Oe s}^{-1}$). The range of field is from 0 to 1.2 Oe and a narrower range (0–0.5 Oe) is selected for investigating the DW motion in figure 2(c) for the reasons given in the first paragraph of section 3.

The average grain sizes of the annealed polycrystalline specimens are small and innumerable DWs exist, so statistical averaging is inevitably necessary to interpret the observed results. We therefore adopt the concept of a 'magnetic object' (MO) (Bertotti 1986) which is a group of neighbouring DWs correlated with each other, equivalent to a

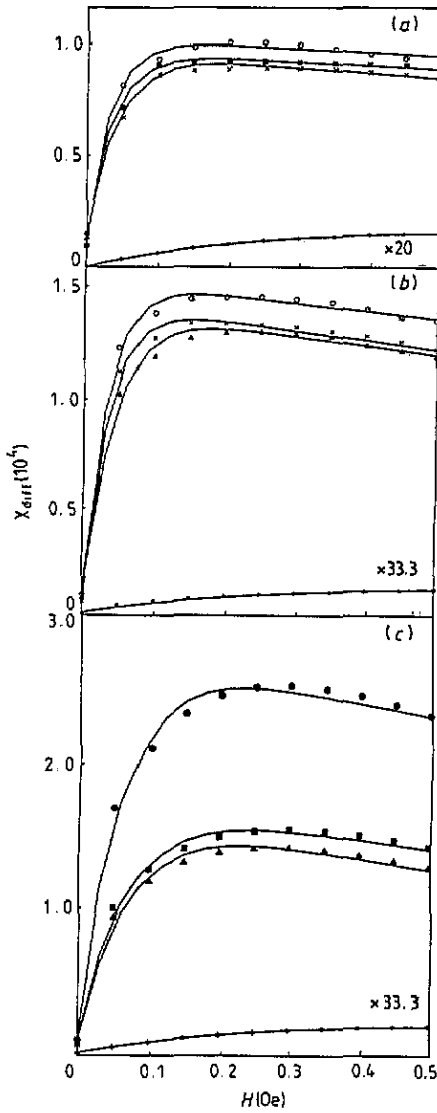


Figure 2. Susceptibility χ_{diff} as a function of magnetic field at different sweep rates for (a) Ni wire, (b) Permalloy-42 wire and (c) 4% Si-Fe sheet (\bullet , $\dot{H} = 43 \text{ mOe s}^{-1}$; \circ , $\dot{H} = 62 \text{ mOe s}^{-1}$; \blacksquare , $\dot{H} = 67 \text{ mOe s}^{-1}$; \times , $\dot{H} = 72 \text{ mOe s}^{-1}$; \blacktriangle , $\dot{H} = 75 \text{ mOe s}^{-1}$ and $*$, $\dot{H} = 6.5 \text{ Oe s}^{-1}$). The full curves are fitted following the parametrization of (1) and they do not pass through all the data points, showing that the assumption that the DW motion predominates in this field range only holds approximately for the wire and sheet samples.

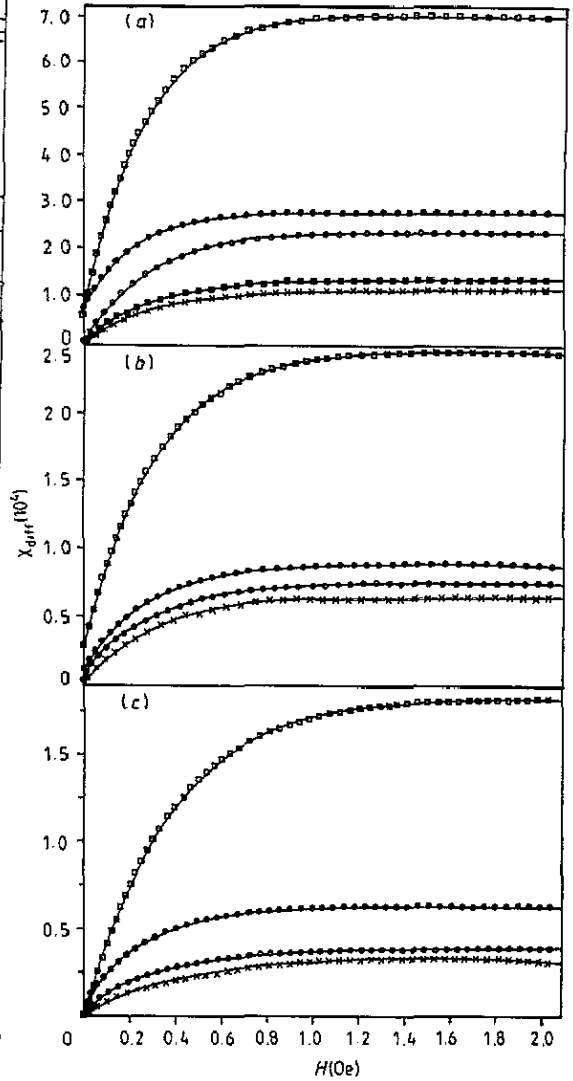


Figure 3. Susceptibility χ_{diff} as a function of magnetic field at different sweep rates for (a) pure Fe bar; (b) 1015 steel bar and (c) 1045 steel bar (\square , $\dot{H} = 15 \text{ mOe s}^{-1}$; \bullet , $\dot{H} = 43 \text{ Oe s}^{-1}$; \circ , $\dot{H} = 62 \text{ mOe s}^{-1}$; \blacksquare , $\dot{H} = 67 \text{ mOe s}^{-1}$; \times , $\dot{H} = 75 \text{ mOe s}^{-1}$). The $\chi_{\text{diff}}-H$ curve at $\dot{H} = 72 \text{ mOe s}^{-1}$ is not plotted owing to congestion, as well as the curves at $\dot{H} = 67 \text{ mOe s}^{-1}$ in (b) and (c).

single object in the sense of a dynamic property to provide coherent magnetization changes along the length of the wire or bar. Each MO contributes independently. DWs, the moving units in magnetization, are thus enlarged to MOs in a polycrystalline sample and supposed

to follow the same equation of motion, i.e. a DW is used in the sense of an MO hereafter.

χ_{diff} , the change of magnetization per unit field, at the early stage of magnetization is related to the total number of DWs which change states of motion in that field region. A phenomenological model has to consider contributions of all changed DWs to χ_{diff} . As discussed in II, different DW energy states are determined by various factors, such as the crystallite orientation and the distribution of internal stress and defects, in a complicated way. At any instant, there are DWs starting and continuing to move, or stopping. DWs in annealed soft ferromagnetic materials can move in very low magnetic fields, e.g. at $H > 0.05$ Oe a large number of DWs in a 3% Si-Fe picture frame have non-zero velocities (Stewart 1951). DW mobilities in these materials are very high, especially in wires, e.g. of the order of $10000 \text{ cm s}^{-1} \text{ Oe}^{-1}$ in low fields (DeBlois 1958). On the other hand, DWs are inevitably stopped because of the finite size of the cross section of the samples, the defect pinning and recombination and disappearance, etc. However, change of DW motion states occurs at different instants owing to the different energy states of the DWs. II shows that mechanisms for moving and stopping are different, so the number of moving DWs varies in a different way from that of stopped DWs. The same parametrization as that of the dynamic internal friction is now applied to the field dependence of the differential susceptibility. This is the difference of two terms, each of which assumes a field exponent. Assume that at a value H of the field which is larger than H_0 , a lower limit for extensive DW motion (\sim tens of mOe for soft ferromagnetic materials), the positive contribution of moving DWs and the negative contribution of stopping DWs are $c_1 \exp(-b_1 H)$ and $c_2 \exp(-b_2 H)$ respectively and hence

$$\chi_{\text{diff}} = c_1 \exp(-b_1 H) - c_2 \exp(-b_2 H) \quad (1)$$

where c_1 , c_2 , b_1 and b_2 are four parameters. This expression reduces to the Rayleigh law at low fields with the approximation $\exp(-bH) \simeq 1 - bH$:

$$\chi_{\text{diff}} = (c_1 - c_2) + (c_2 b_2 - c_1 b_1) H. \quad (2)$$

So $4\pi(c_1 - c_2)$ is the initial permeability and $4\pi(c_2 b_2 - c_1 b_1)$ the Rayleigh constant if we adhere to the approximation $B \simeq 4\pi M$.

The software MATLAB is used to fit the results with a tolerance of ~ 0.01 as shown in figures 2 and 3. The fitted curves are plotted as full curves and the fitted parameters are listed in table 1. It is readily seen that the fitted curves do not pass through all the data points in figure 2, showing the limitation of the assumption of the DW motion for the wire and sheet samples. The deviation could result from a field range which is too large to take DW motion as the only mechanism in the magnetization process, or from samples whose size is too small to contain a large enough number of MOs to produce an average signal following the deduction based on statistics. The parameters c_1 and c_2 are associated, respectively, with the number of moving and stopped DWs (multiplied by a factor K/NSH where $K = k \times 10^8/4\pi$ and k is a proportionality constant) and they are similar in magnitude. $c_1 \geq c_2$ always holds. In wire and sheet samples, c_1 , c_2 and $c_1 - c_2$ decrease from Si-Fe to Permalloy to Ni. In the bar samples, they decrease from pure Fe to 1015 steel to 1045 steel. In general, they all decrease with field sweep rate. On the other hand, the decay constants b_1 and b_2 are different by at least an order of magnitude at low sweep rates. In the wire and sheet samples, b_1 increases from Ni to Si-Fe but b_2 follows the opposite trend. In the bar samples, $b_{1,2}$ are about one order of magnitude smaller than those in the wire (sheet) samples and fluctuation in both constants is obvious. These data are consistent with the following facts:

(i) the number of DWs in Si-Fe sheet taking part in the magnetization process is larger than that in Ni wire but the DWs of Si-Fe sheet are more strongly pinned; the number in Permalloy wire is in between;

(ii) the number of DWs in bar samples taking part in magnetization processes, both moving and stationary, is much larger than that in wire and sheet samples owing to the larger volume;

(iii) the number of moving or stationary DWs (in the sense of MOS) decays much faster in the wire and sheet samples than in the bar samples, which shows the limitation imposed by the relatively large wire surface;

(iv) the number of DWs taking part in the magnetization process decreases when the impurity content increases and especially when the sweep rate becomes larger; and

(v) in wire or sheet samples, the number of moving DWs decreases faster with increasing sweep rate but that of stationary DWs decreases more slowly; in bar samples, the trend is masked by fluctuations.

Table 1. Parameters c_1 , c_2 , b_1 , b_2 and $M_s(\text{DW})$.

Sample	\dot{H} (mOe s)	c_1 (10^4)	c_2 (10^4)	b_1 (Oe^{-1})	b_2 (Oe^{-1})	$M(\text{DW})$ (10^4 G)
Si-Fe sheet	43.0	2.82	2.69	0.381	16.43	1.12
	67.0	1.80	1.67	0.475	14.16	0.68
	75.0	1.75	1.65	0.630	13.77	0.63
	6500.0	0.137	0.071	1.172	2.485	0.032
Permalloy wire	62.0	1.54	1.44	0.253	30.24	0.68
	72.0	1.44	1.37	0.322	29.60	0.62
	75.0	1.41	1.32	0.324	23.74	0.60
	6500.0	0.018	0.017	0.373	3.441	0.004
Ni wire	62.0	1.03	0.910	0.161	31.15	0.47
	67.0	0.976	0.821	0.181	27.36	0.44
	72.0	0.974	0.820	0.272	22.95	0.42
	6500.0	0.071	0.071	1.188	2.179	0.005
Fe bar	15.0	7.16	6.53	0.0142	3.682	12.34
	43.0	2.86	2.15	0.0101	4.429	5.18
	62.0	2.47	2.47	0.0170	3.319	4.11
	67.0	1.43	1.43	0.0170	3.201	2.37
	72.0	1.43	1.43	0.0175	3.112	2.35
	75.0	1.19	1.19	0.0180	3.016	1.94
1015 steel bar	15.0	2.63	2.31	0.0165	3.113	4.43
	43.0	0.955	0.796	0.0145	3.313	1.64
	62.0	0.796	0.716	0.0154	3.315	1.35
	67.0	0.716	0.589	0.0132	3.678	1.25
	72.0	0.716	0.676	0.0166	3.106	1.19
	75.0	0.716	0.660	0.0151	3.433	1.22
1045 steel bar	15.0	1.91	1.91	0.0202	2.533	2.99
	43.0	0.653	0.573	0.0142	3.552	1.12
	62.0	0.406	0.374	0.0170	2.921	0.671
	67.0	0.342	0.326	0.0206	2.182	0.523
	72.0	0.350	0.342	0.0196	2.471	0.549
	75.0	0.366	0.326	0.0146	2.435	0.589

These facts show that the moving and stopping mechanisms are different, while being affected by field and field sweep rate as well as by sample geometry. In the magnetization

process, the moving DWs always predominate but the stopped DWs become important when the sweep rate increases to the order of units of Oe s⁻¹. A slow field sweep rate makes it possible for DWs to move sufficiently so that χ_{diff} is larger.

Based on equation (1), the magnetization is

$$M = (c_1/b_1)[1 - \exp(-b_1 H)] - (c_2/b_2)[1 - \exp(-b_2 H)] \quad (3)$$

satisfying $M = 0$ at $H = 0$. If H tends to the knee, the area below the $\chi_{\text{diff}}-H$ curve is just $M(\text{DW})$, the contribution of DW motion to the saturation magnetization M_s at the early stage of magnetization. The values of $M(\text{DW})$ are also listed in table 1. $M(\text{DW})$ of the wire and sheet samples are about 60% of the published values. They decrease from Si-Fe to Permalloy to Ni and also decrease with sweep rate. $M(\text{DW})$ of the bar samples is larger than the published values, especially at the lowest sweep rate. In general, they decrease from Fe to 1015 steel to 1045 steel and also decrease with the sweep rate. Values of the saturation magnetization obtained by the ballistic method are smaller because of the much faster rate of change of the field. The sweep rate dependence of $M(\text{DW})$ is investigated from the data. The best estimate based on these limited data gives $M(\text{DW}) \propto 1/\dot{H}$ except for the Si-Fe sheet for which $M(\text{DW}) \propto 1/\dot{H}^{0.7}$. It is necessary to determine whether this reciprocal dependence of $M(\text{DW})$ on \dot{H} holds for other soft ferromagnetic materials with geometries different from cylindrical. Similarly, more data are necessary for ascertaining whether $\chi_{\text{diff}} \propto 1/\dot{H}$ in general.

We can estimate the DW velocity based on equation (10(a)) of II:

$$dx/dt = (l\dot{H}/M_s)\chi_{\text{diff}} \quad (4)$$

where l is the distance of the DW equilibrium position from the crystal surface and M_s the saturation magnetization. Taking l as the maximum crystallite size (for the MO, the diameter of the cross section), ~ 1 mm. $\dot{H} \simeq 10^{-2}$ Oe s⁻¹; $M_s \simeq 10^4$ Oe and $\chi_{\text{diff}} \simeq 10^4$. The DW velocity is hence estimated as 10^{-3} cm s⁻¹ which is one order of magnitude smaller than 0.017 cm s⁻¹ in the Fe-Si picture-frame single crystal (Williams *et al* 1950). This reflects the fact that the hindrances to DW motion in polycrystalline materials are more serious than in a single crystal.

The wire samples of Ni and Permalloy-42 were the same as those used in experiments on internal friction under magnetization (the pure Fe wire could not be used owing to weak induction signals). We can compare the parameters $b_{1,2}$ of χ_{diff} with their counterparts in the dynamic internal friction Q_m^{-1} (table 2 in II): b_1 is similar in magnitude but b_2 of χ_{diff} is at least one order of magnitude larger. This shows that χ_{diff} is also affected by applied alternating stress: the stopping of DWs slows down owing to extra energy gained in the stress field. Hence, although the same form of parametrization is applicable in both cases, the IF $Q_m^{-1}-H$ relations are not simply related to the $\chi_{\text{diff}}-H$ curves by a proportional constant.

4. Summary

To describe the relationship of χ_{diff} versus H in smoothly increasing fields, we propose a parametrization expression with four parameters $c_{1,2}$ and $b_{1,2}$. Change of DW motion states under field sweeping can hence be studied. A very low-rate field sweeping can sufficiently increase the number of moving DWs, which increases χ_{diff} and the contribution of DW motion to magnetization. However, high sweep rates lower both χ_{diff} and the saturation magnetization. The data also show the influences of sample geometry and impurity.

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