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FMR study of surface and isothermal annealing effects in amorphous $Fe_{80}B_{20-x}C_x$ and $Fe_{82}B_{18-y}Ge_y$ alloys

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Alterations in the bulk (surface) magnetic properties of amorphous $Fe_{80}B_{20-x}C_x$ ($0 \leqslant x \leqslant 10$) and $Fe_{82}B_{18-y}Ge_y$ ($0 \leqslant y \leqslant 6$) alloys, caused by isothermal annealing (mechanical polishing) of the alloy ribbons at a temperature $T_A = 0.5T_{cr}$ (where T_{cr} is the crystallization temperature) for various lengths of time, t_a, ranging from 0 to 240 min, have been studied by measuring the corresponding changes in the ferromagnetic resonance (FMR) linewidth, ΔH_{pp}, and resonance field, H_{res}. Polishing induces significant changes in both H_{res} and ΔH_{pp} but leaves their composition dependence practically unaltered. By comparison, isothermal annealing has no influence on H_{res} for all the investigated alloy compositions whereas it affects ΔH_{pp} to different extents depending on the alloy composition. Physical implications of these results are discussed in terms of the effect of surface and annealing treatments on various contributions to H_{res} and ΔH_{pp} in the glassy alloys in question.

1. Introduction

Amorphous alloys are generally prepared by rapid quenching either from the liquid or vapour state. Since an infinite cooling rate cannot be achieved in practice, the structure of amorphous alloys differs not only from that of the melt but also from the "ideal" amorphous structure because of large quenched-in local stresses that originate from sudden and non-uniform cooling. Materials prepared in this way are, therefore, thermodynamically unstable and subsequent annealing at moderate temperatures causes structural relaxation – a process leading to a more stable amorphous state rather than to the most stable ordered (crystalline) state. Structural relaxation (SR) in metallic glasses induces changes in a large number of physical properties [1]. The extent of change depends upon the degree of sensitivity of a given property to slight modifications in structure. For instance, small but finite changes are observed in density and radial distribution function (RDF) whereas the properties that get drastically affected by SR include mechanical embrittlement, atomic diffusivities, internal friction, electrical resistivity and magnetic properties. Changes in some of the above mentioned properties, e.g. density, diffusivity and RDF, are manifestations of the irreversible relaxation phenomena and basically result from the increased topological atomic short-range order (TSRO), which is defined, in contrast with the compositional short-range order (CSRO), as the short-range order in the atomic positions regardless of the chemical identity of individual atoms and is caused by the collective atomic movements rather than the diffusion of individual atoms. Another interesting aspect of low-temperature annealing is the reversible changes such a treatment produces in field-induced magnetic anisotropy, Curie temperature and resistivity. Reversible relaxation phenomena of this type are essentially brought about by alterations in CSRO. Changes in the physical properties as a consequence of annealing at moderate temperatures, therefore, provide a powerful means of studying the relaxation processes. Investigation of structural relaxation processes, in turn, is vital to the understanding of thermal stability and structure of metallic glasses. One of the most informative tools to study SR particularly in ferromagnetic amorphous alloys is the ferromagnetic resonance (FMR) technique [2] because FMR linewidth is extremely sensitive to changes in the local magnetic anisotropy and the internal fields induced by SR (or by some other means).

Detailed X-band FMR measurements on amorphous $Fe_{80}B_{20-x}C_x$ ($0 \leqslant x \leqslant 10$) and $Fe_{82}B_{18-y}Ge_y$ ($0 \leqslant y \leqslant 6$) alloys have been performed in order to gain physical insight into the mechanisms of structural relaxation, caused by isothermal annealing at $T_A = 0.5T_{cr}$ (where T_{cr} is the crystallization temperature in °C) for various lengths of time, t_a, ranging from 0 to 240 min, in these systems. Choice of the above mentioned alloy series for the type of study intended was motivated by the unique possibility these systems offer to investigate the effects, if any, of the CSRO in the un-annealed state on the phenomenon of structural relaxation. To elucidate this point further, in the "as-quenched" state, the CSRO in the carbon substituted glassy alloys differs markedly from that in the germanium substituted ones because the replacement of B by C in a-$Fe_{80}B_{20-x}C_x$ alloys occurs at the interstitial position whereas that of B by Ge in a-$Fe_{82}B_{18-y}Ge_y$ alloys takes place at the substitutional position on account of the difference in

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the metalloid atomic radii [3, 4]. The physical implications of the observed variations of FMR peak-to-peak linewidth (ΔH_{pp}) and resonance field (H_{res}) with composition for "as-received" (un-annealed and unpolished) and polished alloy ribbons and with the annealed time for the unpolished samples are discussed in terms of the following well-known [5–7] expressions for H_{res} and ΔH_{pp}, obtained by solving the Landau–Lifshitz–Gilbert (LLG) equation along with Maxwell's equations for the parallel geometry used in the present measurements, i.e.

$$(\omega/\gamma)^2 = (H_{res} - H_{dem} + H_k) \times (H_{res} - H_{dem} + H_k + 4\pi M_s) \qquad (1)$$

and

$$\Delta H_{LLG} = 1.45\lambda\omega/\gamma^2 M_s \qquad (2)$$

where H_{dem} is the demagnetizing field, H_k is the in-plane uniaxial anisotropy field, M_s is the saturation magnetization, $\gamma = -g|e|/2mc$ is the gyromagnetic ratio, λ is the Gilbert damping parameter and $\nu = \omega/2\pi$ is the frequency of the microwave field, and the observation [5, 6, 8–10], based on the FMR measurements performed on a wide variety of amorphous ferromagnetic alloys at constant temperature but at different frequencies of the microwave field, that besides the LLG contribution, which varies linearly with ν, there exists an additional contribution, ΔH_l, to ΔH_{pp}, which does not depend on ν and arises from the inhomogeneity in the internal field mainly caused by exchange fluctuations, inhomogeneous local magnetization and local random anisotropy.

2. Experimental details

Amorphous (a-) $Fe_{80}B_{20-x}C_x$ ($x = 0, 2, 4, 6, 8, 10$) and $Fe_{82}B_{18-y}Ge_y$ ($y = 0, 2, 4, 6$) alloy ribbons of ~ 1 mm width and $\sim 30\,\mu$m thickness were prepared by the melt spinning technique and procured from General Electric Company, New York. The crystallization temperature, T_{cr}, values for different compositions were determined from the differential scanning calorimetry (DSC) scans recorded at a heating rate of $10°C\,min^{-1}$ on the Perkin-Elmer DSC-4 differential scanning calorimeter. 3 mm long strips cut from the alloy ribbons were isothermally annealed at $T_A = 0.5T_{cr}$ in nitrogen atmosphere for various durations of time, t_a, ranging from 15 to 240 min. Nitrogen, as an exchange gas, ensures that the sample attains the annealing temperature in less than 10 min. Figure 1 depicts schematically the thermal treatment received by the samples prior to the measurement. Samples taken out of the furnace as soon as they attain the annealing temperature, T_A, (i.e. $t_a = 0$ and $t_r \simeq 10$ min in Fig. 1) served as "reference" for investigating the type and extent of structural relaxation that had taken place during the time intervals t_r and t_a by comparing the data on these reference samples with those obtained on the corresponding un-annealed samples and on the samples that had undergone isothermal annealing at T_A for different lengths of time, t_a, respectively. Fresh ribbon strip, from the same batch, was taken for isothermal annealing every time the annealing time was different because for every subsequent annealing treatment of the previously annealed sample the reference time (t_r) adds up to the annealing time, t_a, but during t_r the sample temperature is not steady at T_A. In order to make sure that there is no initiation of crystallization in the annealed ribbons, they were examined for crystalline regions after every annealing treatment under a scanning electron microscope. First derivative FMR absorption spectra were recorded at room temperature ($\simeq 300$ K) in the X-band (operating frequency $\simeq 9.3$ GHz) on JEOL FE-3X EPR spectrometer before and after the samples had undergone annealing or surface treatments, using the horizontal-parallel geometry in which the external magnetic field (H_a) is applied along the length in the ribbon plane to minimize demagnetization effects. Preliminary FMR data on a-$Fe_{82}B_{18-y}Ge_y$ have been reported elsewhere [11] but the detailed results and analysis are presented in this work.

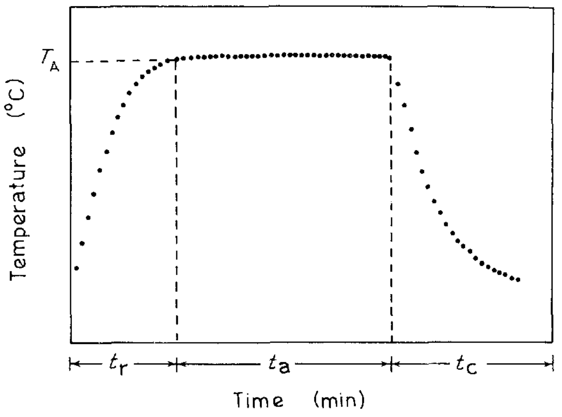

Figure 1 Schematic representation of the thermal treatment received by the samples prior to FMR measurement. T_A, t_r, t_a and t_c denote the annealing temperature, reference time $\simeq$ 10 min, annealing time and cooling time $\simeq$ 10 min, respectively.

3. Results and discussion

3.1. Crystallization temperature

DSC scans recorded at a heating rate of $10°C\,min^{-1}$ for the glassy alloys in question are depicted in Figs 2 and 3. It is noticed from these figures that the Ge-substituted glasses exhibit a broad and shallow exothermic peak which at higher temperature is followed by another much sharper and intense exothermic peak as contrasted with the amorphous $Fe_{80}B_{20-x}C_x$ alloys and the alloy with $y = 0$ in the amorphous $Fe_{82}B_{18-y}Ge_y$ alloy series for which the DSC scans show a single but sharp exothermic peak. In keeping with the current understanding of crystallization in metallic glasses [12], the shallow peak in Ge-substituted alloys results from the primary crystallization of α-Fe phase, which coexists with the amorphous Fe–B–Ge matrix, which, in turn, transforms at high temperatures (sharp peak in DSC scans) by the polymorphous crystallization into an ordered stable $Fe_3(B, Ge)$ crystalline compound, whereas the single sharp peak in the remaining glassy alloys arises on account of eutectic crystallization in which two crystalline phases α-Fe and $Fe_3(B, C)$ simultaneously

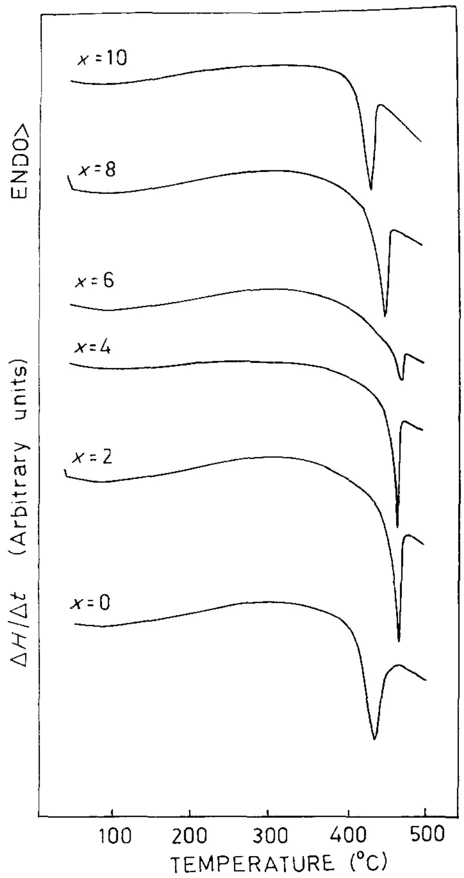

Figure 2 DSC scans for amorphous $Fe_{80}B_{20-x}C_x$ alloys. Scan rate = $10° min^{-1}$.

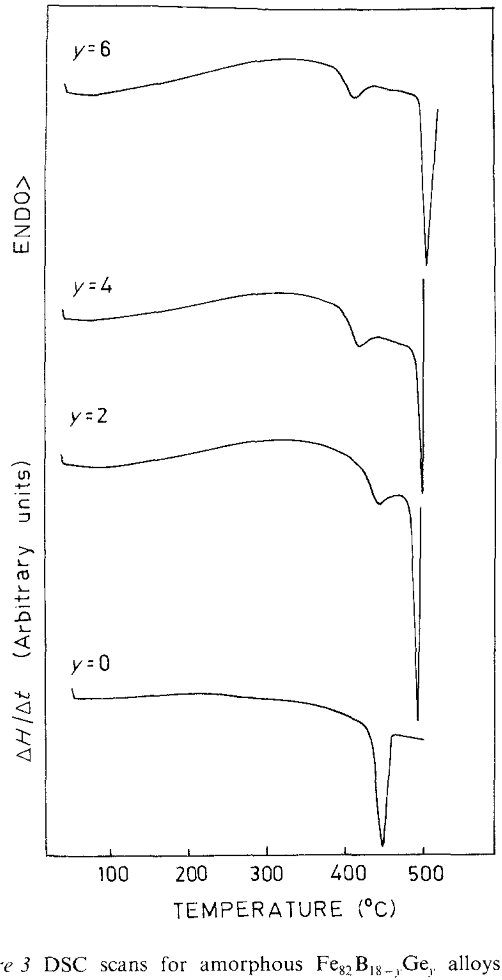

Figure 3 DSC scans for amorphous $Fe_{82}B_{18-y}Ge_y$ alloys. Scan rate = $10° min^{-1}$.

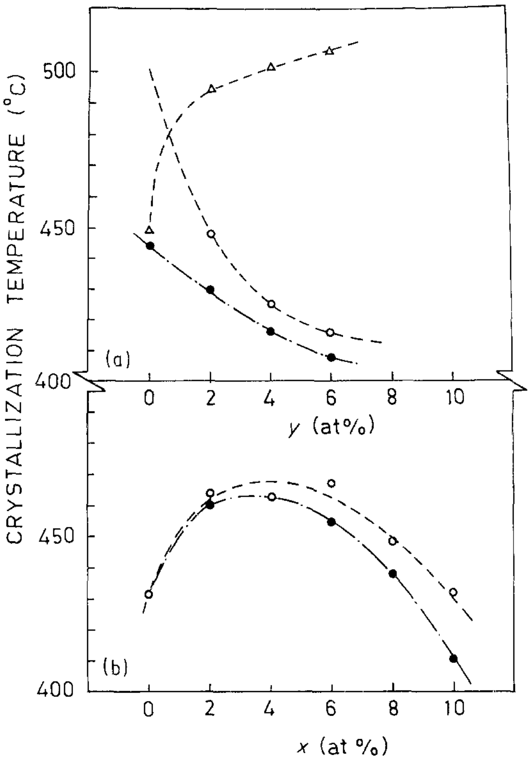

Figure 4 Composition dependence of the crystallization temperature for amorphous (a) $Fe_{80}B_{20-x}C_x$ and (b) $Fe_{82}B_{18-y}Ge_y$ alloys. (a) (●) Ref. [4]; (○, △) present work, (b) (●) Ref. [3]; (○) present work.

crystallize out. Temperatures corresponding to various DSC peaks are taken to represent the crystallization temperatures, T_{cr}. Concentration dependence of T_{cr} for the two glassy alloy series is shown in Fig. 4. In this figure, the crystallization temperatures, determined by Luborsky *et al.* [3, 4] from the thermomagnetic curves taken at a fixed heating rate of $20° C min^{-1}$ on the same alloys as the present ones, have been included for comparison. Since T_{cr} values given by Luborsky *et al.* [3, 4] are actually the temperatures marking the onset of crystallization and manifesting themselves in the beginning of an increase in magnetization, their values lie consistently lower than those determined in this work for a-$Fe_{80}B_{20-x}C_x$ alloys despite slower heating rate used by us. In the case of a-$Fe_{82}B_{18-y}Ge_y$ alloys, the eutectic crystallization temperature for a-$Fe_{82}B_{18}$ and primary crystallization temperatures for the alloys with $y = 2$, 4 and 6 are reasonably close to the T_{cr} values determined by Luborsky and Walter [4]. However, Luborsky and Walter [4], for some reason, have failed to detect the polymorphous crystallization temperature for the alloys with $y = 2$, 4 and 6, which are considerably higher than the primary crystallization temperatures. Contrary to the conclusions drawn by Luborsky and Walter [4], the present investigations coupled with earlier crystallization studies on Fe–B–Si amorphous alloys [13] demonstrate that the crystallization behaviour (and hence the thermal stability) of Fe–B–Ge and Fe–B–Si amorphous alloys is markedly similar but differs from that of the glassy Fe–B–C alloys.

3.2. Effect of polishing

Surface composition and structure are expected to

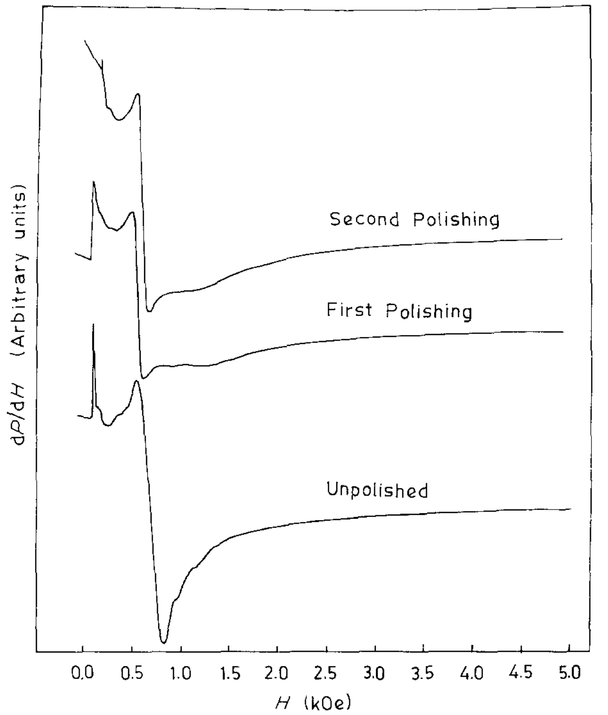

Figure 5 Typical FMR power absorption derivative curves for unpolished, "first-time-polished" and "second-time-polished" alloy ribbons.

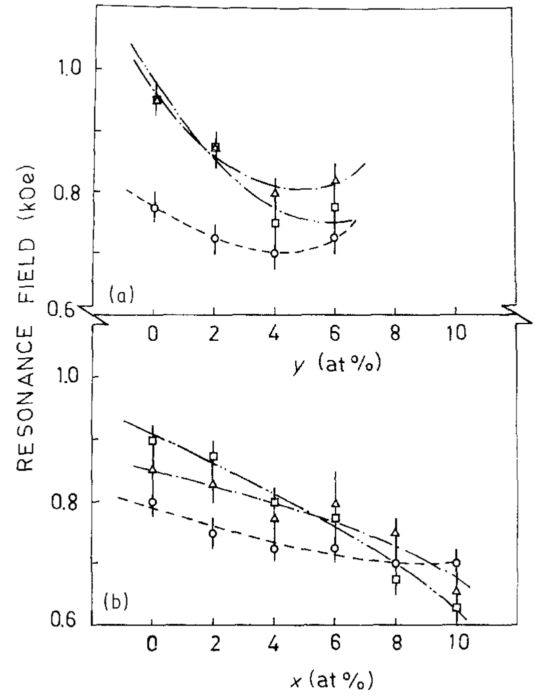

Figure 6 Effect of polishing on the composition dependence of resonance field. (UP) unpolished; (1P) "first-time-polished"; (2P) "second-time-polished" samples (a) $Fe_{82}B_{18-y}Ge_y$; (b) $Fe_{80}B_{20-x}C_x$. (○) UP; (△) 1P; (□) 2P.

have significant influence on the resonance field, H_{res}, and the peak-to-peak FMR linewidth, ΔH_{pp}, because the "skin-depth" penetration of the microwaves is confined to a very thin surface layer. A rough calculation presented below shows that the thickness of this surface layer does not exceed 1 μm. For the investigated glassy alloys, the typical value of electrical resistivity at room temperature is $\simeq 120\,\mu\Omega$ cm [14, 15] and that of μ ($\omega \simeq 58.5$ GHz) lies in the range 100 to 10. Using these values in the expression for the skin depth, i.e. $\delta = c/[4\pi\sigma\omega\mu(\omega)]^{1/2}$, where $\omega = 2\pi\nu$ ($\nu \simeq 9.3$ GHz), σ is the specific d.c. electrical conductivity, c is the velocity of light and μ is the frequency-dependent permeability, gives $\delta \simeq 0.1$ to 1 μm as compared with the typical thickness 30–40 μm of the alloy ribbons. In order to assess the magnitude of change in the values of H_{res} and ΔH_{pp} caused by surface effects, FMR power absorption derivatives (PAD) at $\nu = 9.3$ GHz have been measured at room temperature ($\simeq 300$ K) in the horizontal parallel configuration in the static magnetic field range of 0 to 5000 Oe before and after the "as-received" sample had been mechanically polished using 600 grade emery paper and cleaned in acetone by the ultrasonic method. After recording the PAD curves on the samples that had been polished once (1P), the same samples were polished again (2P) before taking FMR spectra on them. The typical results obtained on the ("as-received") unpolished (UP), 1P and 2P samples are shown in Fig. 5 whereas Figs 6 and 7 serve to illustrate the effect of polishing on H_{res} and ΔH_{pp}. H_{res} and ΔH_{pp} both are noticed (Figs 6 and 7) to increase upon polishing by 10 to 25% for a-$Fe_{80}B_{20-x}C_x$ (with the exception of the alloy with $x = 10$, for which both H_{res} and ΔH_{pp} decrease by about 10%) and a-$Fe_{82}B_{18-y}Ge_y$ alloys. Before embarking upon the interpretation of the changes in H_{res} and ΔH_{pp} induced by polishing, it is imperative to ascertain whether Equations 1 and 2 are capable of explaining the magnitude of these quantities for "as-received" (un-annealed and unpolished) alloy samples.

Table I lists the values of H_{res} for "as-received" ribbons of a-$Fe_{80}B_{20-x}C_x$ and a-$Fe_{82}B_{18-y}Ge_y$ alloys calculated using $g = 2.09 \pm 0.02$ [5, 6, 8–10, 16] and the room temperature values [3, 4] of $4\pi M_s$ in Equation 1 when (i) both H_{dem} and H_k in Equation 1 are neglected and (ii) only the anisotropy term is dropped and values of H_{dem}, computed from the actual dimensions of the investigated alloy ribbons, are inserted in Equation 1. A comparison between the values of H_{res}, so calculated, and those observed experimentally (Table I) demonstrate that the former values for the case (ii) are in striking agreement with the latter ones (within the error limits) for all the glassy alloys in question except

TABLE I Observed and calculated values of the resonance field for the amorphous $Fe_{80}B_{20-x}C_x$ and $Fe_{82}B_{18-y}Ge_y$ alloys in the "as-received" condition; H^1_{res} and H^2_{res} are the values of H_{res} computed from Equation 1 when (1) both H_{dem} and H_k are ignored and (2) only H_k in Equation 1 is dropped

Alloy	M_s (G)	H_{dem} (Oe)	Calculated		Observed H_{res} (Oe)
			H^1_{res} (Oe)	H^2_{res} (Oe)	
$x = 0$	1337.6	173.1	623.2	746.8	800 ± 25
$x = 2$	1336.6	173.0	623.6	747.0	750 ± 25
$x = 4$	1329.9	172.2	626.5	748.9	725 ± 25
$x = 6$	1314.8	170.2	633.2	753.1	725 ± 25
$x = 8$	1318.2	170.6	631.7	752.1	750 ± 50
$x = 10$	1333.0	172.6	625.2	748.0	700 ± 25
$y = 0$	1326.2	171.7	628.1	749.8	750 ± 25
$y = 2$	1299.2	168.2	640.2	757.6	725 ± 50
$y = 4$	1354.6	175.3	615.9	742.2	750 ± 25
$y = 6$	1370.6	177.4	609.2	738.1	725 ± 25

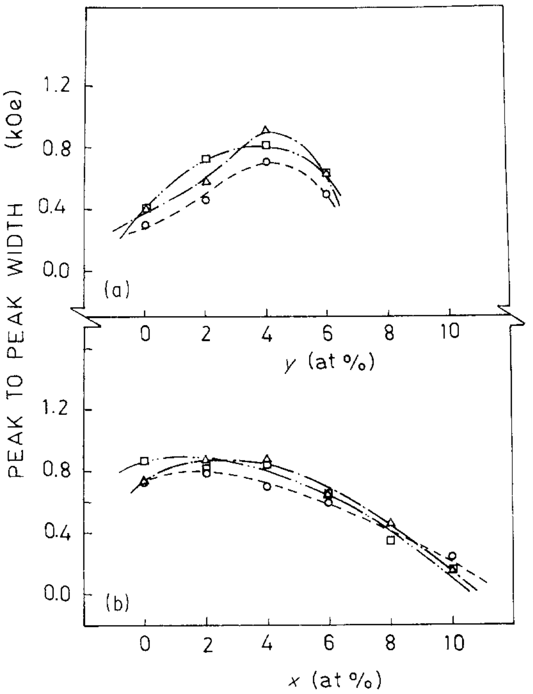

Figure 7 Effect of polishing on the composition dependence of peak-to-peak linewidth. (a) $Fe_{82}B_{18-y}Ge_y$; (b) $Fe_{80}B_{20-x}C_x$. (○) UP; (△) 1P; (□) 2P.

for a-$Fe_{80}B_{20}$ and a-$Fe_{80}B_{10}C_{10}$. Such an agreement (discrepancy) between calculated and observed values of H_{res} implies that a total neglect of the in-plane uniaxial anisotropy is justified (not justified). If the magnetization vector lies in the sample plane along the length because of the uniaxial anisotropy, the anisotropy term in Equation 1 has a positive sign. The reported value [17] of uniaxial anisotropy constant $K_u \simeq 3 \times 10^4\,\mathrm{erg\,cm^{-3}}$ for a-$Fe_{80}B_{20}$ yields $H_k = 2K_u/M_s \simeq 45\,\mathrm{Oe}$, substituting this value in Equation 1 and solving for H_{res} gives $H_{res} = 702\,\mathrm{Oe}$, which is not only considerably smaller than the observed value, 800 Oe, but also lower than the value $\simeq 750\,\mathrm{Oe}$ obtained by taking into account the demagnetizing field only. By contrast, if this anisotropy causes the magnetization vector to point along the breadth in the sample plane, the anisotropy term in Equation 1 has a negative sign. If H_k is taken to be negative in Equation 1 and the above value of $H_k = 45\,\mathrm{Oe}$ is used, H_{res} comes out to be 792 Oe, a value in very good agreement with the observed value. Similarly, if the sign of H_k for a-$Fe_{80}B_{10}C_{10}$ is positive and the same value of H_k as that for a-$Fe_{80}B_{20}$ is used in Equation 1, H_{res} has a value $\simeq 703\,\mathrm{Oe}$ very close to that ($700 \pm 25\,\mathrm{Oe}$) observed. The above inference that the direction of the magnetic anisotropy, though in the ribbon plane, does not coincide with the ribbon axes (ribbon length) for a-$Fe_{80}B_{20}$ is in conformity with the observed domain patterns and Mössbauer spectra [17] on this material. As contrasted with a quantitative comparison between theory and experiment in the case of H_{res}, discussion of ΔH_{pp} is bound to remain at a qualitative level only since we do not have facilities needed to carry out FMR measurements at different frequencies. Frequency dependent FMR data reported [5, 6, 8–10] on a number of widely different amorphous ferromagnetic alloy systems and commercial metallic glasses reveal that $\Delta H_l \simeq (0.6\text{–}0.7)\,\Delta H_{pp}$ ($\nu = 9.3\,\mathrm{GHz}$) and ΔH_{LLG} contributes the rest (i.e. $\Delta H_{LLG}(\nu = 9.3\,\mathrm{GHz}) \simeq (0.3\text{–}0.4)\,\Delta H_{pp}(\nu = 9.3\,\mathrm{GHz})$). This observation will be used while discussing the change in ΔH_{pp} caused by either polishing or annealing treatments.

M_s, H_{dem} and g, being bulk properties, are not expected to get affected by surface treatments such as mechanical polishing so that, in view of Equation 1 and the arguments presented above, the increase (decrease) in H_{res} due to polishing results from the fact that the strains introduced in the surface tilt the easy axis in the ribbon plane away from the ribbon axis through the magnetostrictive effect (the easy axis continues to point along the length in the ribbon plane but the surface value of anisotropy constant increases) but this effect is confined to the surface only. Again, polishing cannot have any influence on the microscopic inhomogeneities and hence on ΔH_l (this is also clearly borne out by other experiments [6]), the observed change in ΔH_{pp} should, therefore, be caused by the corresponding change in ΔH_{LLG}. According to Equation 2, the only quantity in the expression for ΔH_{LLG} that can alter on account of surface treatment is the Gilbert damping parameter λ. Consistent with earlier observations [6], the present data indicate a considerable increase in the surface value of λ induced by polishing for all the investigated glassy alloys except for the a-$Fe_{80}B_{10}C_{10}$ alloy, in which case $\lambda_{surface}$ decreases. These deductions retain their validity even when the contribution to ΔH_{pp} resulting from the skin depth effect (which makes the magnetization induced by the microwave field non-uniform in the volume of the surface penetration layer) is also taken into account because this contribution for the glassy alloys of the type presently investigated turns out to be negligibly small, as is shown by a straightforward calculation based on the expression [18] for the linewidth due to skin effect relevant to the present case ($H_a \ll 4\pi M_s$), i.e. $\Delta H = (16\pi/3)(A\sigma\omega)^{1/2}/c$, where $A = JS^2M_sa^2/g\mu_B$ is the exchange stiffness parameter, J is the exchange coupling constant, S and a are the spin and lattice parameter, respectively. Using the typical values $J \simeq 3\,\mathrm{meV}$ [19, 20], $a \simeq 0.354\,\mathrm{nm}$, M_s (300 K) [21] $\simeq 1.9\mu_B/a^3$, σ (300 K) [14, 15] $\simeq 0.94 \times 10^{17}\,\mathrm{sec}^{-1}$, $S = 1$ [19, 20], $g = 2.09$ [5, 6, 8–10, 16], $\nu = \omega/2\pi \simeq 9.3\,\mathrm{GHz}$ in the above expression yields $\Delta H \simeq 15\,\mathrm{Oe}$. Even if polishing causes a 100% change in this contribution, altered value of ΔH would still lie well within the error limits.

3.3. Effect of isothermal annealing

Changes in the FMR PAD curves representative of the glassy alloy systems in question and caused by isothermal annealing (on unpolished samples) at $T_A = 0.5\,T_{cr}$ during time intervals ranging from 15 to 240 min are illustrated by Fig. 8. Variation of H_{res} and ΔH_{pp} with annealing time, t_a, for different compositions in the amorphous alloy series $Fe_{80}B_{20-x}C_x$ and $Fe_{82}B_{18-y}Ge_y$ is depicted in Figs 9 to 12. The salient features of the results presented in these figures are: (i) isothermal annealing for $t_a \leqslant 240\,\mathrm{min}$ does not have any influence (within the error limits) on H_{res}

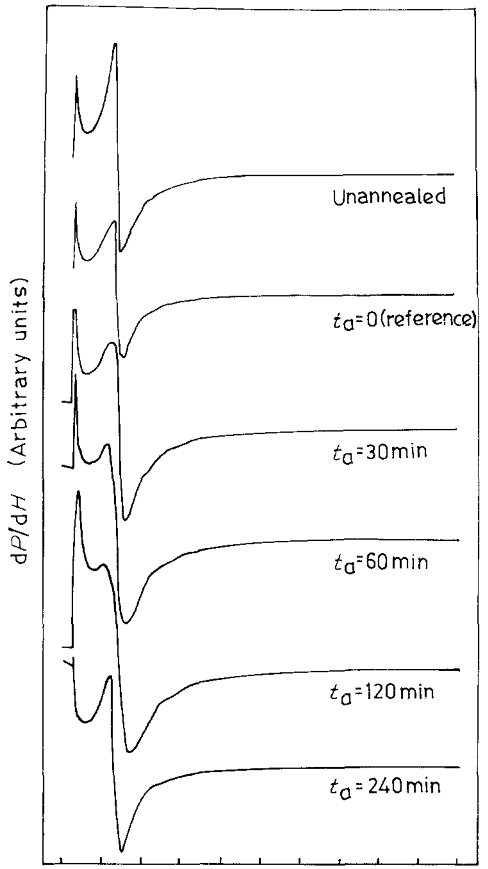

Figure 8 Typical room temperature FMR spectra recorded at various stages of isothermal annealing on a given unpolished sample.

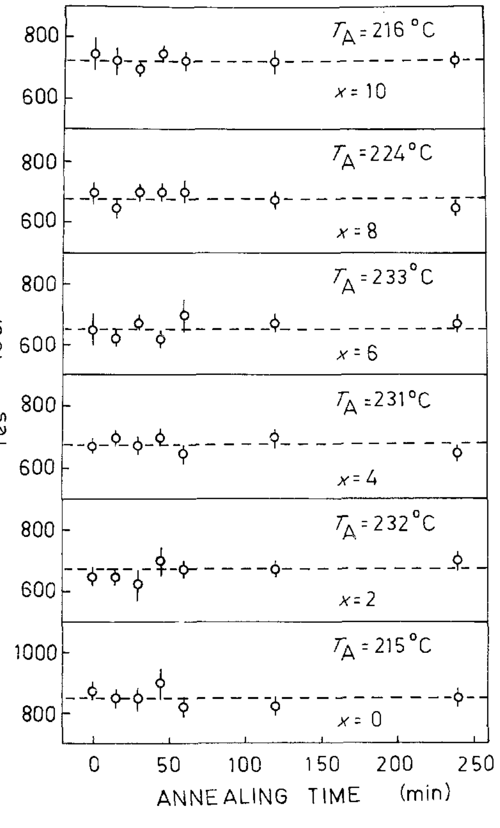

Figure 9 H_{res} as a function of annealing time, t_a, for amorphous $Fe_{80}B_{20-x}C_x$ alloys.

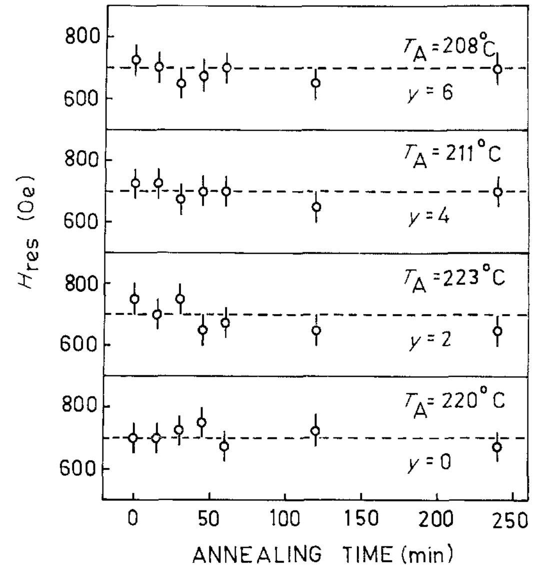

Figure 10 Variation of the resonance field with annealing time for amorphous $Fe_{82}B_{18-y}Ge_y$ alloys.

for all the glassy alloy compositions studied in this work, and (ii) ΔH_{pp} does not exhibit any variation with t_a (i.e. stays constant within the uncertainty limits) for the compositions $x = 0, 2, 4, 10$ and $y = 2$ in the amorphous alloy series $Fe_{80}B_{20-x}C_x$ and $Fe_{82}B_{18-y}Ge_y$, respectively, but for the remaining alloys the functional dependence of ΔH_{pp} on t_a differs from alloy to alloy, e.g. for the alloys with $x = 6$ and $y = 6$, $\Delta H_{pp}(t_a) = \Delta H_{pp}(0) + bt_a$; for $y = 4$, $\Delta H_{pp}(t_a) = \Delta H_{pp}(0) - b't_a$; ΔH_{pp} initially increases and then saturates for $t_a \geq 120$ min for $x = 8$ and ΔH_{pp} as a function of t_a goes through a broad peak around $t_a \simeq 120$ min for $y = 0$. Comparison between the results obtained on the "virgin" as-quenched and "reference" samples reveals that the annealing process, in which the annealing temperature evolves with time and attains the value T_A at $t = t_r$ (Fig. 1), leaves H_{res} unaltered for all the investigated alloy compositions, does not change ΔH_{pp} from its value for the "virgin" samples for the alloys with $x = 0, 2, 4, 10$ and $y = 0, 6$ but decreases ΔH_{pp} by about 20 to 25% for the remaining alloys ($x = 6$, 8 and $y = 2, 4$).

In the light of the arguments presented in the preceding section and Equation 1, the observation that H_{res} retains its "virgin" state value despite isothermal (or "dynamic" for $t \leq t_r$) annealing treatment suggests that the isothermal annealing has no effect on M_s (and hence on H_{dem}), a deduction in consonance with the similar observation made by Heinrich *et al.* [8] on a-$Fe_{100-x}B_x$ alloys. This inference coupled with the general finding [22, 23] that the Gilbert damping parameter, λ, is directly proportional to M_s implies that λ is also not affected by isothermal annealing. This result, in agreement with the outcome of previous studies [6, 8], strongly indicates that the LLG contribution to linewidth; ΔH_{LLG}, (Equation 2) does not change with annealing so that the observed variation of ΔH_{pp} with t_a is entirely due to the change in ΔH_i. Moreover, inhomogeneous local magnetization and anisotropy field (both of dipolar and magnetostrictive origin) and exchange fluctuations in the glassy materials under consideration give rise to a

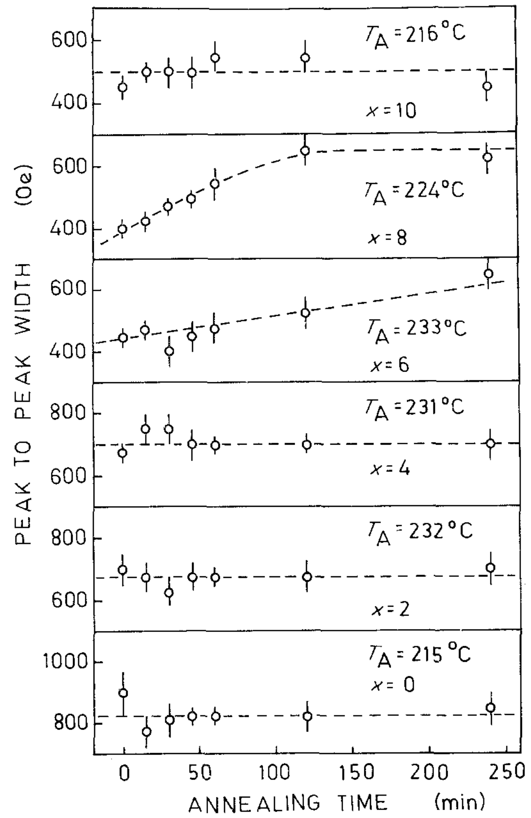

Figure 11 Variation of the peak-to-peak linewidth with annealing time for various compositions in the alloy series a-Fe$_{80}$B$_{20-x}$C$_x$.

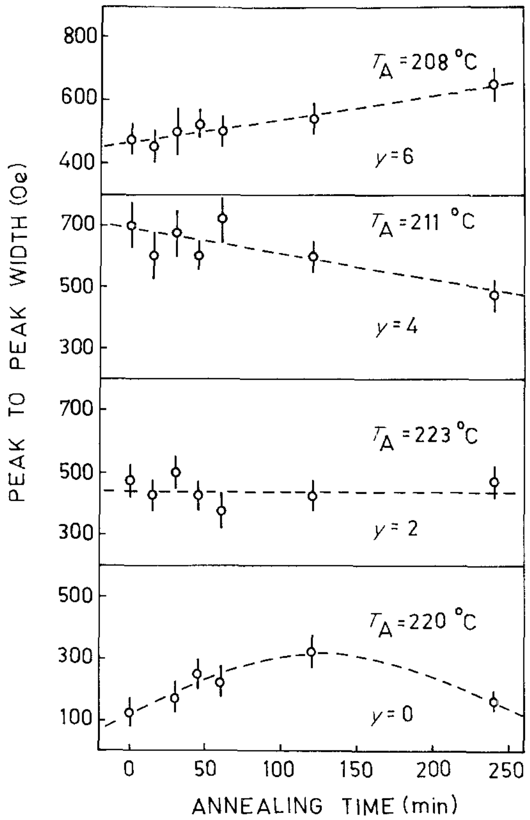

Figure 12 Peak-to-peak linewidth as a function of annealing time for different compositions in the amorphous alloy series Fe$_{82}$B$_{18-y}$Ge$_y$.

contribution (ΔH_l) to linewidth which accounts for 70% of the total linewidth (ΔH_{pp}) observed at ν = 9.3 GHz (see Section 3.2). Now that the values of the in-plane uniaxial anisotropy field, H_k, do not exceed 45 Oe in presence of saturation magnetizations ($4\pi M_s$) of about 15 kG, a value of 45 Oe for H_k when used in the relevant expression given in [23] yields a maximum linewidth anisotropy broadening of about 30 Oe, which lies within the error limits of our data. Also, the contribution to ΔH_l due to exchange fluctuations cannot exceed that arising from the exchange conductivity mechanism, i.e. $\Delta H \simeq$ 15 Oe (skin effect). Thus, the contribution originating from inhomogeneous local magnetostrictive anisotropy field dominates over other contributions to ΔH_l. That this term could be of sizeable magnitude is also supported by the finding that these materials possess both large magnetostriction and large local quenched-in stresses. Stress relief occurring during the annealing process is, therefore, expected to have considerable influence on this contribution to ΔH_l. Consequently, the decrease (increase) of ΔH_{pp} with t_a can be attributed to the re-orientation of spins in the glassy ribbons through a magnetostrictive effect towards a lower energy configuration which corresponds to a more homogeneous (inhomogeneous) distribution of magnetic moments than in the "virgin" sample as contrasted with no change in ΔH_{pp} for certain alloys implying that the spins in the "virgin" sample are already in the minimum energy configuration and annealing at intermediate temperatures does not alter ΔH_l and hence ΔH_{pp}. However, a clear-cut understanding of why there is no systematic trend of the relaxation behaviour with composition is still lacking. A possible explanation could be that the different alloys have relaxed (structurally) to different extents due to room temperature annealing for about three years (time gap between preparation and the present measurements) but this conjecture is not supported by the crystallization temperature against concentration data (Fig. 4) which demonstrate a close agreement between not only the presently determined T_{cr} values and those previously reported [3, 4] on the samples coming from the same batch as ours (provided our values for the onset of crystallization are compared with those reported in [3, 4]) but also between their composition dependence. Finally, since the functional dependence of ΔH_{pp} on t_a is not distinctly different in the two amorphous alloy series, CSRO in the un-annealed state does not seem to influence the structural relaxation consequent upon annealing at intermediate temperatures in any significant way.

4. Conclusions

The present data permit us to draw the following conclusions:

1. The crystallization behaviour (and hence the thermal stability) of Fe–B–Ge and Fe–B–Si amorphous alloys is markedly similar but differs from that in the glassy Fe–B–C alloys.

2. Mechanical polishing tilts the easy axis in the ribbon plane away from the ribbon axis and increases the LLG damping parameter, λ, by about 10 to 25% in a surface layer of about 1 μm thickness for all the investigated alloys with the exception of the alloy a-Fe$_{80}$B$_{10}$C$_{10}$ for which polishing increases the surface value of the anisotropy (dipolar plus magnetostrictive) constant without changing the direction of easy axis and decreases λ by about 10%.

3. LLG damping in the bulk and saturation magnetization remain unaltered by isothermal annealing treatment.

4. Stress relief during the isothermal annealing process results in the re-orientation of spins in these glassy alloys through a magnetostrictive effect towards a lower energy configuration and hence affects the short-range magnetic order in them.

5. CSRO in the un-annealed state does not cause any noticeable change in the structural relaxation consequent upon isothermal annealing.

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