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Experimental observation of quantum corrections to electrical resistivity in nanocrystalline soft magnetic alloys*

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Abstract. X-ray diffraction patterns of nanocrystalline Fe–Cu–Nb–Si–B (FINEMET) alloys reveal that bcc α-Fe/α-FeSi crystallites with the average grain size of 20(5) nm are dispersed in amorphous matrix. Enhanced electron–electron interaction (EEI) and quantum interference (QI) effects as well as electron–magnon (and/or electron-spin fluctuation) scattering turn out to be the main mechanisms that govern the temperature dependence of resistivity. Of all the inelastic scattering processes, inelastic electron–phonon scattering is the most effective mechanism to destroy phase coherence of electron wave functions. The diffusion constant, density of states at the Fermi level and the inelastic scattering time have been estimated, for the first time, for the alloys in question.

Keywords. Fe–Cu–Nb–Si–B; soft magnetic alloys; nanocrystalline; resistivity; quantum interference effects; electron–magnon scattering; electron-spin fluctuation scattering.

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1. Introduction

A critical survey of the literature on nanocrystalline materials reveals that the structural, mechanical/elastic, thermal and magnetic attributes have been extensively studied. The main reason for this appears to be the increased mechanical strength, improved ductility, higher thermal expansion coefficient and superior soft magnetic properties in comparison with conventional coarse grained materials. Other fundamental properties such as electrical, thermal and galvanomagnetic transport have received little, or even no, attention. Many fundamental changes in the magnetization processes have been observed when a nanocrystalline magnetic material is compared with its crystalline counterpart. Due to

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the sizable magnetic contribution, transport properties are also expected to reflect such changes. Now that the electron mean-free path is expected to be extremely small in such materials, electrical resistivity provides a *local* probe to study alterations in the local atomic structure brought about by the partial crystallization of the amorphous matrix leading to the formation of nanocrystalline materials. Taking cognizance of these arguments, a detailed investigation of the temperature dependence of electrical resistivity of nanocrystalline Fe–Cu–Nb–Si–B soft magnetic alloys (commercially known as FINEMET) over a wide temperature range was undertaken.

2. Experimental details

Fe-based nanocrystalline alloys with composition $Fe_{74.78}Cu_{0.92}Nb_{2.63}Si_{15.3}B_{6.3}$, $Fe_{74.37}Cu_{0.95}Nb_{3.02}Si_{14.15}B_{7.5}$ and $Fe_{73.28}Cu_{0.94}Nb_{2.87}Si_{15.45}B_{7.36}$ (henceforth referred to as S1, S2 and S3, respectively) were produced by *controlled* partial crystallization of 'melt-quenched' amorphous ribbons of cross-section $\sim 2 \times 0.03$ mm^2. A detailed analysis of X-ray diffraction patterns taken at room temperature using Cu K_α radiation revealed that bcc α-Fe/α-FeSi crystallites with average grain size of 20 ± 5 nm are dispersed in the amorphous matrix. Electrical resistivity measurements were performed on the nanocrystalline samples S1, S2 and S3 (typical dimensions $35 \times 2 \times 0.03$ mm^3) in the temperature range 13 K $\leq T \leq$ 300 K at 0.5 K intervals using the four-probe dc method. At every setting, temperature was stabilized to within ± 10 mK and the sample current (typical value 3.5 mA) was reversed to correct the measured voltage for the spurious thermo emf. A relative accuracy of better than 10 ppm was achieved in the present experiments.

3. Results and discussion

The resistivity (ρ) data, shown as the normalized resistivity, $r(T) = \rho(T)/\rho$ (273.15 K), vs. temperature plots for the samples S1, S2 and S3 in figure 1 exhibit minima at a temperature $T_{\min} \approx 100$ K, reminiscent of those observed earlier [1–3] in amorphous magnetic alloys of similar composition. Figures 2 and 3 serve to demonstrate that the theoretical variations (continuous curves) based on the expressions $r(T) = r(0) - aT^{1/2} + bT^2$ (13 K $\leq T \leq T^*$) and $r(T) = r'(0) - a'T^{3/2} + b'T^2$ ($T^{**} \leq T \leq 300$ K) closely reproduce the observed temperature dependence (open circles) in the specified ranges with the upper limit of the low-temperature range $T^* = 34$, 32 and 37 K, the lower limit of the high-temperature range $T^{**} = 90$, 54 and 88 K for the samples S1, S2 and S3, respectively. While the second terms in these expressions can respectively be identified with [1–3], the enhanced electron–electron interaction (EEI) and quantum interference (QI) contributions to $r(T)$, the third term ($b = b'$ within error limits) arises from the electron–magnon and/or electron-spin fluctuation scattering. The EEI and QI contributions to $r(T)$ are given by [4,6] $r(T)_{\rm EEI} = -r(0)\rho(0)(1.294/\sqrt{2})(e^2/3\pi^2\hbar)(k_{\rm B}T/\hbar D)^{1/2}$ and $r(T)_{\rm QI} = -r'(0)\rho(0)(e^2/2\pi^2\hbar)(D\tau_{\rm ie})^{-1/2}$, where D is the diffusion coefficient and $\tau_{\rm ie} = \tau_{\rm ie}^0 T^{-3}$ for $T < \Theta_{\rm D}$ (Debye temperature) is the inelastic scattering time when electron–phonon scattering is the dominant inelastic scattering mechanism. Equating the coefficients of $T^{1/2}$ and $T^{3/2}$ in the above expressions, we obtain the values $D \approx 5$ cm^2/s

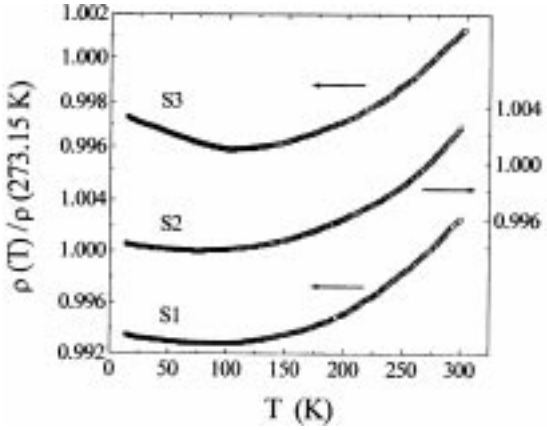

Figure 1. Temperature (T) dependence of normalized resistivity, $\rho(T)/\rho$(273.15 K), for the samples S1, S2 and S3.

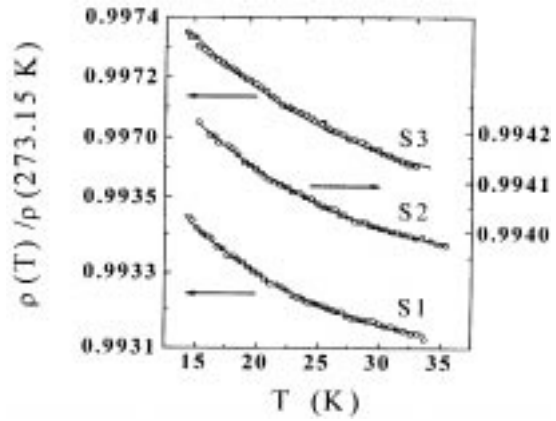

Figure 2. Temperature (T) dependence of normalized resistivity, $\rho(T)/\rho$(273.15 K), for the samples S1, S2 and S3 in the low-temperature region with the theoretical fit (curve).

and τ_{ie} (300 K) $\approx 5 \times 10^{-15}$ s, *irrespective* of the alloys composition. Using the Einstein relation $N(E_F) = [\rho(0)e^2D]^{-1}$, the density of states at Fermi level, $N(E_F) \approx 0.1$ eV^{-1} atom^{-1}. The elastic scattering time τ_{el} computed from the relation $\tau_{el} = m/ne^2\rho(0)$ (where n is the number of electrons per m^3), when compared with τ_{ie} (300 K), asserts that $\tau_{el} \ll \tau_{ie}$. This implies that the inelastic mean free path is much larger than the elastic mean free path in the entire temperature range covered in the present experiments. Thus, it is not surprising that QI effects are important even at temperatures as high as 300 K in the present case.

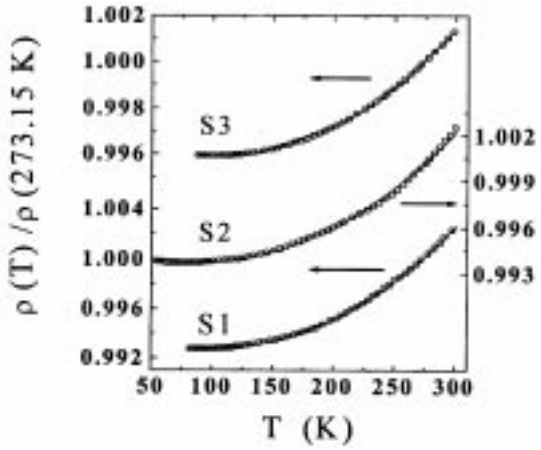

Figure 3. Temperature (T) dependence of normalized resistivity, $\rho(T)/\rho(273.15\ \mathrm{K})$, for the samples S1, S2 and S3 in the high-temperature region with the theoretical fit (curve).

4. Conclusion

The main conclusions are: (1) Enhanced electron–electron interaction effects account for the dominant $-\sqrt{T}$ dependence of $r(T)$ for $T < T_{\min}$. (2) Coherent electron–magnon plus electron-spin fluctuation scattering together with quantum interference effects govern the temperature dependence of resistivity for $T > T_{\min}$. (3) Out of the inelastic scattering mechanisms that destroy phase coherence, electron–phonon scattering is the most effective dephasing mechanism and its dephasing action persists to temperatures as high as 300 K. (4) Diffusion constant, density of states at the Fermi level and the inelastic scattering time have been determined, for the first time, for the nanocrystalline alloys in question.

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