Transport of heat by spin waves in $Fe_{95}Si_5$

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The thermal and electrical conductivities of the alloy $Fe_{95}Si_5$ have been measured between 1.5 and 4 K, in fixed magnetic fields of value 0, 0.22, and 5.5 T. The thermal conductivity decreases with increasing field above saturation, indicating a contribution of spin waves. This contribution reaches 8% at 4 K. The lifetime of thermal spin waves varies as $T^{-1.2}$, in approximate agreement with theoretical predictions for magnon-electron scattering.

I. INTRODUCTION

Transport of heat by thermal spin waves across a temperature gradient was first detected in yttrium iron garnet and in other magnetic insulators.[1-3] A magnetic field is used to remove the spin waves, allowing a separation of the spin-wave contribution from the contribution of lattice waves.

In the case of pure metals, the electronic contribution is too large and too field dependent for such an experiment to be possible. By using concentrated alloys, we have succeeded[4,5] in isolating the spin-wave contribution. The strong scattering of electrons by alloy disorder makes the electronic contribution small, and almost independent of field above ferromagnetic saturation. Using electrical-resistivity measurements, even the small remaining field variation of the electronic contribution can be taken into account.

The collision time of thermal spin waves can be derived from these experiments. For processes involving one spin wave only, this collision time is equal to the spin-wave lifetime τ_m. In the case of nickel-rich Ni-Fe, we found[4] $\tau_m \propto T^{-0.6}$ or T^{-1}. The spin-wave contribution was about 3% of the total thermal conductivity at 4 K. In the case of $Fe_{68}Co_{32}$ it was about 10%, allowing a more accurate determination of τ_m. We found[5] $\tau_m \propto T^{-1.2}$ between 1.5 and 4.5 K. Since the average frequency $\overline{\omega}$ of thermal spin waves is itself proportional to T, these data imply $\tau_m \propto (\overline{\omega})^{-1.2}$. This is close to the relation $\tau_m \propto (\overline{\omega})^{-1}$ characteristic of Gilbert (i.e., Landau-Lifshitz) damping.

The relatively large spin-wave contribution observed in $Fe_{68}Co_{32}$ can be ascribed[5] to an unusually low electronic density of states, making magnon-electron collisions less frequent. Another approach to the choice of a suitable material might be based on the idea of minimizing the electronic contribution. This means alloys with very large electrical resistivity ρ, such as the Fe-Si described in this paper.

II. SAMPLE PREPARATION AND EXPERIMENTAL APPARATUS

The sample was made by levitation melting in an induction furnace, from high-purity Johnson-Matthey iron and silicon. The helium atmosphere present during melting was purified by passage through titanium turnings at 850 °C and through a charcoal trap cooled with liquid nitrogen. To avoid solute segregation and blowholes, the ingot was cast into a horizontal copper boat. After being enclosed in a tight-fitting jacket of soft steel, it was swaged. Then it was machined into the shape of a cylinder 5.3 mm in diameter and 99 mm long. It was annealed for 96 h at 1260 °C in a flow of pure hydrogen dried by passage through a liquid-nitrogen trap. The temperature was then lowered to 900 °C, and the sample was annealed in a vacuum of about 10^{-4} Torr for 4 h in order to remove any dissolved hydrogen. Finally, it was furnace cooled.

Grains of about 3 mm are visible. Wet chemical analysis indicates 2.35-wt.% Si, corresponding to the atomic composition $Fe_{954}Si_{46}$. Spectrographic analysis did not show the presence of any other elements.

The apparatus used for thermal-conductivity measurements was described in Refs. 5 and 6. The electrical heaters are wound directly around the sample. The distance between the two main thermometers attached to the sample is 20 mm. The method used to measure the electrical resistivity was described in Ref. 4, and the electrical current is about 3 A.

III. EXPERIMENTAL RESULTS

The electrical resistivity ρ in zero field is 39.3 $\times 10^{-8}$ Ωm at room temperature and 28.8×10^{-8}Ω m at 4.2 K. In longitudinal fields, it decreases by only 0.17% up to saturation, at 4.2 K. This decrease reflects the existence of a small, negative, "ferromagnetic anisotropy of resistance," caused[7] by spin-orbit interaction. Above saturation, ρ was

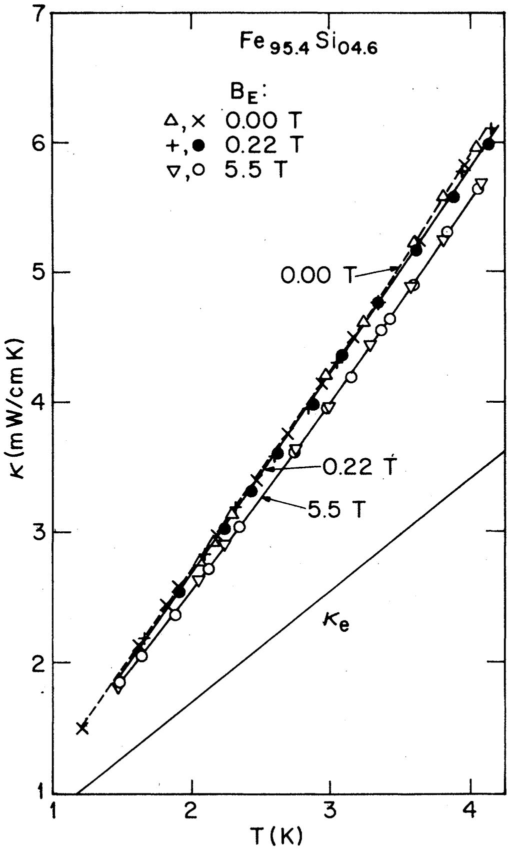

FIG. 1. Thermal conductivity κ of $Fe_{95}Si_5$ as a function of temperature at three different values of the external field B_E. The electronic contribution to κ is denoted by κ_e.

measured at temperatures of 4.2, 1.95, and 1.2 K, and was found to decrease by only ≃0.04% when the external field B_E is increased from 0.22 to 5.5 T. This decrease is comparable to the experimental accuracy of ±0.03% for ρ. For a given value of B_E, ρ is the same at all three temperatures, showing that the alloy is in the residual resistance region. Hence we can use the theoretical Sommerfeld value $L_{th} = 2.44 \times 10^{-8}$ V^2/K^2 of the Lorenz number, in order to calculate the electronic part κ_e by the Wiedemann-Franz law $\kappa_e = L_{th}T/\rho$ (Fig. 1). The small electronic variation $\Delta\kappa_e$ between two fields can be calculated similarly (see Table I).

The thermal conductivity κ was measured at external fields of 0, 0.22, and 5.5 T, all parallel to the heat current (Fig. 1). The last two values are sufficient to saturate the sample. For clarity, only two runs are shown for each field value. Data analysis was described in detail in Ref. 5. The measured variation $\Delta\kappa$ between 0.22 and 5.5 T, as well as the spin-wave variation $\Delta\kappa_m$, are given in Table I. The following equations are used:

$$\Delta\kappa_m = \Delta\kappa - \Delta\kappa_e\,, \quad \kappa_m(T) = 11.883\Delta\kappa_m/\Delta J_D\,, \tag{1}$$

where ΔJ_D is the variation between 0.22 and 5.5 T of a certain function $J_D(g\mu_B M_s/k_BT, g\mu_B B_E/k_BT)$. This function describes[5] the progressive destruction of the spin-wave contribution by the external field B_E. Once the zero-field spin-wave contribution $\kappa_m(T)$ has been obtained from Eqs. (1), the average spin-wave lifetime $\overline{\tau}_m(T)$ is calculated by

$$\kappa_m(T) = 11.883k_B^{5/2}T^{3/2}/16\pi^2\hbar D^{1/2}\alpha\,, \qquad \overline{\tau}_m = 1/2\alpha\overline{\omega}\,, \quad \overline{\omega} = 1.8k_BT/\hbar\,, \tag{2}$$

where α is the Gilbert damping constant, and $\overline{\omega}$ an appropriate average[5] of frequencies of thermal spin waves. We assume $M_s = 2.10$ T (Ref. 8), $g = 2.10$ (Ref. 9). The exchange stiffness constant is $D \simeq 265$ meV Å^2 $= 4.2 \times 10^{-40}$ J m^2 (Ref. 10). We assume[5] the "dirty" limit $\Lambda_e q \ll 1$ to hold in our analysis, where Λ_e is the electron mean free path and q is the spin-wave wave number.

Typical values of q would be $4.8 \times 10^{+8}$ m^{-1} at 4.0 K and $3.0 \times 10^{+8}$ m^{-1} at 1.5 K, in zero field. If we introduce the actual Lorenz number $L = \kappa\rho/T$ for our alloy, our data at 4 K give $L/L_{th} = 1.7$.

IV. DISCUSSION OF RESULTS

Our results (Table I) show that spin waves contribute about 8% of the heat conduction at 4 K. Figure 2 indicates that κ_m varies like $T^{1.3}$ on the average between 1.5 and 4 K. Correspondingly, we find $\overline{\tau}_m \propto T^{-1.2}$ in the same temperature range (Fig. 2). Since $\overline{\omega} \propto T$ [see Eqs. (2)], this implies $\overline{\tau}_m \propto (\overline{\omega})^{-1.2}$. This leads, in turn, to an almost constant value for the Gilbert parameter (Table I).

These results are similar to those obtained[5] for $Fe_{68}Co_{32}$ (Fig. 2) and already mentioned in Sec. I. A constant Gilbert parameter can be explained in

TABLE I. The fifth column shows the spin-wave thermal conductivity. The sixth column shows the Gilbert parameter. The seventh and eighth columns show the average frequency and average lifetime of thermal magnons.

T (K)	$\Delta\kappa$ (mW/cm K)	$\Delta\kappa_e$ (mW/cm K)	$\Delta\kappa_m$ (mW/cm K)	κ_m (mW/cm K)	α (10^{-3})	$\overline{f} = \overline{\omega}/2\pi$ (GHz)	$\overline{\tau}_m$ (10^{-10} sec)
1.5	−0.0662	0.0005	−0.0667	0.141	3.19	56.4	4.39
2.0	−0.113	0.0007	−0.114	0.214	3.23	75.2	3.25
2.5	−0.163	0.0008	−0.164	0.291	3.33	94	2.52
3.0	−0.213	0.0010	−0.214	0.374	3.41	113	2.05
3.5	−0.256	0.0011	−0.257	0.449	3.57	132	1.68
4.0	−0.272	0.0013	−0.273	0.485	4.04	150	1.30

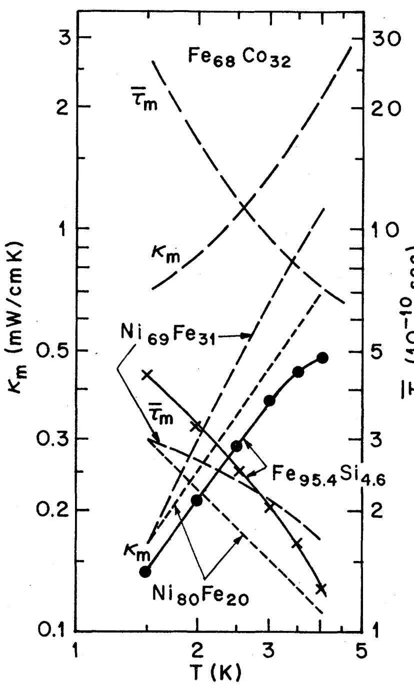

FIG. 2. Spin-wave contribution κ_m to thermal conductivity as a function of temperature at zero field. Also, average spin-wave lifetime $\bar{\tau}_m$ as a function of temperature. The values of κ_m and $\bar{\tau}_m$ are shown for various alloys, including our $Fe_{95}Si_5$.

terms of magnon-electron scattering.[11] In one type of collision process,[12] a magnon is absorbed or emitted while an itinerant electron is scattered between the spin-up and spin-down Fermi surfaces. This "spin-flip" process is mediated by the isotropic s-d exchange interaction. Spin-lattice relaxation is fast enough in alloys to keep the spins of itinerant electrons in an equilibrium distribution, despite this influx of angular momentum from the magnons. In a second type of collision,[12] the electron is scattered between two points of the same Fermi surface, while a magnon is absorbed or created. Since the total spin is not conserved, a combination of exchange and spin-orbit interactions is needed to activate such "nonflip" processes. This combination can be expressed[13] as a simple spin Hamiltonian of the anisotropic exchange variety.

Using this spin Hamiltonian formalism, the theory can be cast in a physically transparent form.[13] In the case of nonflip processes, it is very similar to the theory of sound attenuation in metals. For example, the concept of "effective zone" and the parameter $\Lambda_e q$ are important. At $\Lambda_e q \ll 1$, the constancy of the Gilbert parameter is a simple consequence[13] of the Pauli exclusion principle.

Physical insight can also be gained from an interesting recent theoretical work.[14] This paper expresses the magnon scattering probability in the same form of an "overlap integral" between electron spectral density functions [Eq. (4.11) in Ref. 14] as does Ref. 13. The finite electron mean free path Λ_e controls the broadening of these spectral functions in momentum space, thus changing their degree of overlap, and in turn the value of τ_m.

In $Fe_{95}Si_5$ (as well[5] as in $Fe_{68}Co_{32}$), $\bar{\tau}_m$ varies as $T^{-1.2}$ rather than T^{-1}. Correspondingly, Table I shows that α increases by about 25% between 1.5 and 4 K. Perhaps a small amount of Rayleigh scattering of spin waves by alloy disorder ($\bar{\tau}_m \propto q^{-5}$) exists[15] and is superposed on the dominant magnon-electron scattering. Estimates on the basis of Ref. 15 indicate that this Rayleigh scattering may have the right order of magnitude to explain the observed increase of α.

V. VALUES OF THE GILBERT PARAMETER

It is unfortunate that the Landau-Lifshitz parameter $\lambda = \gamma M_s \alpha$, where $\gamma = g\mu_B/\hbar$, has been used so extensively by workers in the field of ferromagnetic resonance. It contains the saturation magnetization M_s in a way that has no microscopic justification, and that leads to incorrect conclusions when materials having different M_s are compared.

The Gilbert parameter α is more meaningful than λ, and has the advantage of being dimensionless. In Table II, we give a survey of λ and α values for various metallic materials, derived from existing ferromagnetic-resonance (FMR) and thermal-conductivity (TC) data. Most of the quoted FMR data have been obtained at frequencies of at least 30 GHz, where the intrinsic linewidth is less affected by experimental errors. It is disturbing to see that the α values measured by FMR in Refs. 18 and 20 for Fe-Si are only one-half of our value, $\alpha \simeq 3.4 \times 10^{-3}$, for $Fe_{95}Si_5$. Note, however, that the α value for $Ni_{57}Fe_{43}$ quoted in the Ref. 18 is also suspiciously smaller than values obtained by other authors[17,19,4] for Ni-Fe alloys.

Not only are the experimental values of α for these various materials somewhat uncertain, but it is not even certain whether the spin-flip or the nonflip mechanism is dominant. In the case of Ni and Ni-Cu, orbital degeneracies and the nonflip mechanism are probably responsible for the large α values.[14] An important factor affecting α in the case of both mechanisms is the electronic density of states, as measured by the electronic specific-heat coefficient γ (Table II). The γ value[21] for our $Fe_{95}Si_5$ differs little from the value for pure iron or for Ni-Fe. Another important factor is the electronic mean free path Λ_e as indicated by the value of the electrical resistivity ρ (see Table II). The relatively large ρ for our $Fe_{95}Si_5$ should have

TABLE II. Values of Landau-Lifshitz parameter λ, of Gilbert parameter α, of electronic specific-heat coefficient γ, and of electrical resistivity ρ. Thermal–conductivity method is denoted by TC, and ferromagnetic resonance by FMR.

Material	Method	Reference	T (K)	λ_{cgs} (10^8 sec^{-1})	α (10^{-3})	γ (mJ/mol K^2)	$\rho(T)$ (10^{-8} Ω m)
Nickel	FMR	16	293	2.3	24.9	7	6.8
Iron	FMR	16	293	0.7	2.2	5	9.7
Cobalt	FMR	16	293	1.0	3.74	5	6.2
$Ni_{75}Fe_{25}$	FMR	17	300	0.88	5.1	4.1	20
$Ni_{57}Fe_{43}$	FMR	18	300	0.42	1.5	4.1	26
$Ni_{69}Fe_{31}$	FMR	19	77		4.41	4.0	6.2
$Ni_{69}Fe_{31}$	TC	4	1.5–4.5		3.1	4.0	4.2
$Ni_{80}Fe_{20}$	TC	4	1.5–4.5		4.65	4.4	4.3
$Ni_{69}Cu_{31}$	TC	4	1.5–4.5		> 26.5	6.5	23.4
$Fe_{68}Co_{32}$	TC	5	1.5–4.5		0.73	1.9	5.5
$Fe_{95}Si_5$	TC	present data	1.5–4.0		3.4	5	28.8
$Fe_{94}Si_6$	FMR	18	300	0.41	1.38	5	47
$Fe_{96}Si_4$	FMR	20	300	0.41	1.35	5	36

the effect of increasing α in the case of the spin-flip mechanism, and to decrease it for the nonflip mechanism. But no clear correlation between ρ and α is visible in Table II. An attempt has been made recently[13,14] to explain this paradox in terms of the complexity of the 3d Fermi surface, where various sheets approach each other arbitrarily closely.

VI. CONCLUSIONS

In $Fe_{95}Si_5$, the spin-wave contribution amounts to 8% of the total thermal conductivity at 4 K. This large contribution allows an accurate determination of the lifetime τ_m of spin waves, which is found to vary as $T^{-1.2}$. This points to magnon-electron scattering as the dominant mechanism of spin-wave relaxation.

Our present results suggest that the thermal-conductivity technique can be used to study spin relaxation in a very wide class of alloys. The only requirement or restriction is for the alloy to have, like $Fe_{95}Si_5$, an electrical resistivity of at least $\simeq 20 \times 10^{-8}$ Ω m below 4 K. The present technique is especially useful and successful in the case of iron-rich alloys, where the ferromagnetic resonance technique is inaccurate because the very narrow intrinsic linewidths are easily masked by exchange conductivity and other effects.

ACKNOWLEDGMENT

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