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On the cross-over from half-metal to normal ferromagnet in NiMnSb

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Abstract. Magnetism and transport properties of the semi-Heusler compound NiMnSb are re-examined in great details. A wide set of experiments (elastic and inelastic neutron scattering, static magnetic measurements, magnetoresistance, Hall effect, thermopower, FMR) have been performed on polycrystals, single crystals or single-crystalline thin films, and the results are analysed. Special emphasis is given to the magnetic excitations and to the relaxation mechanisms in this metallic ferromagnet. At low temperatures, all experimental results hint at the existence of a fully spin-polarized conduction band (half metallic state). At higher temperature ($T > 80$ K), but well below the Curie temperature (730 K), a cross-over to a usual metallic ferromagnetic state is evidenced and discussed.

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1 Introduction

NiMnSb is the first ferromagnet where the occurrence of half-metallic character at low temperature was predicted from band calculations [1]. This feature was proved later experimentally by Hanssen *et al.* [2], by comparing the angular correlations of positron annihilation with theoretical calculations. However, attempts failed to prove the full polarisation of conduction electrons at the Fermi level, either when studying the photoemission on thin films [3] or studying spin valves or tunnel junctions based on NiMnSb [4,5], or when measuring the Andreev reflection at the interface with a superconductor [6]. The probable reason for such failures when using these surface-sensitive techniques is to be found in the atomic disorder in NiMnSb thin films or at the interfaces [7].

However, one can wonder if the half metallic character can be maintained up to the Curie temperature or if the half metallic gap closes before the disappearance of the ferromagnetic band splitting. Most probably NiMnSb should turn to a normal ferromagnet below the Curie point. We present in this paper experimental evidences which suggest that changes in electronic properties happen near 80 K, far below the Curie point of NiMnSb ($T_C = 730$ K) and we try to demonstrate that these phenomena are due to a cross-over from the half metallic phase at lower temperatures to a normal ferromagnet at higher ones.

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2 Previous experimental data

Otto *et al.* [8,9] measured the magnetisation, resistivity, Hall effect on polycrystalline NiMnSb samples in the temperature range 4–800 K. They give a rather complete description and interpretation of the physical properties of this compound, especially concerning the mechanism of the Hall effect. Among many other features, they observed in NiMnSb a rapid rise of the ordinary Hall coefficient R_H^0 when lowering the temperature below 80 K, whereas this coefficient was found almost constant between 100 and 300 K. They also found an anomaly in the ratio of the anomalous Hall effect to the resistivity below 94 K. However, they do not comment these anomalies. Moodera and Mootoo [10] studied the transport properties of thin films. In addition to an anomaly of R_H^0 below 40 K, they found that below 100 K the resistivity ρ dropped under the curve extrapolated from the temperature range 120-300 K. They observed a nearly linear variation of ρ in the range 2–15 K, which they related to the half metallic character.

Hordequin *et al.* [11] re-examined the magnetisation and resistivity of NiMnSb samples including single crystals in the temperature range 1.5–300 K, and the spin wave dispersion relation using neutron inelastic scattering. The spontaneous magnetisation M_s reaches 4.025 ± 0.02 μ_B per formula at 2 K. The excess above the integer value may be due to a slight off-stoichiometry or to a polarisation of internal shells. A very weak magneto- crystalline anisotropy favours the $\langle 110 \rangle$ axes, no change in the easy axis can be detected up to 120 K where the anisotropy

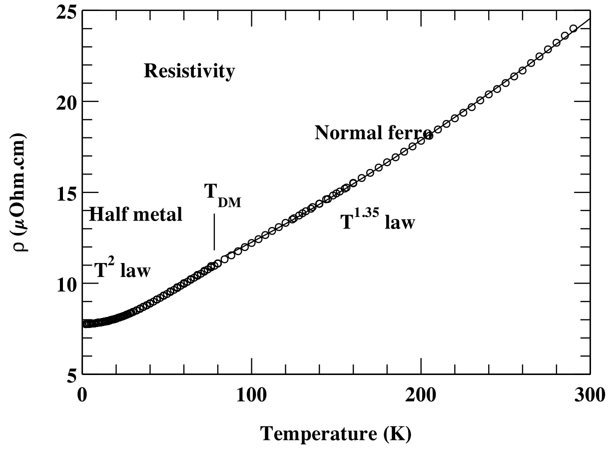


Fig. 1. Resistivity *versus* temperature for a NiMnSb polycrystal.

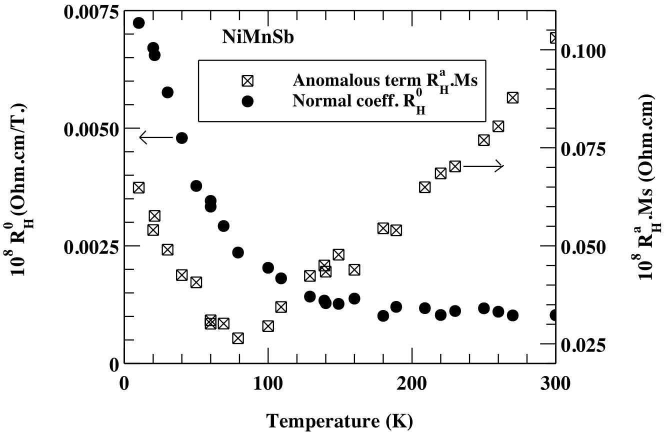


Fig. 2. Normal and anomalous Hall coefficients for NiMnSb.

becomes negligible. The regime of the spontaneous magnetisation M_s – followed along the 3 principal inequivalent crystallographic directions – changes near 70–80 K: below this temperature, $M_s(T)$ behaves as $T^{3/2}$, as it is the case for Heisenberg ferromagnets at low temperatures. Conversely, above 100 K, M_s^2 is a linear function of T^2 up to about 500 K, which is the classical behaviour for itinerant ferromagnets.

A change in the resistivity behaviour was observed near 80 K (Fig. 1). However, contrary to the observations of Moodera and Mootoo, a nearly quadratic behaviour of ρ was observed below 50 K, as well on different bulk samples as on thin films: $\rho = \rho_0 + BT^{\alpha}$, with $1.7 < \alpha < 2.2$. Let us note that the residual resistivity of the studied samples (3 to 20 μohm cm) was much smaller than for the samples of Moodera and Mootoo, which can explain the difference in behaviour. Conversely, between 100 and 300 K, the resistivity follows the law: $\rho = A_2 + B_2 T^{\alpha_2}$, where $1.3 < \alpha_2 < 1.5$ and the value A_2 is larger than the residual resistivity value ρ_0. Let us note that the shape of the resistivity-*versus*-temperature curve, with a negative second derivative near 80 K, cannot be explained by the superposition of a normal spin wave contribution and a phonon Grüneisen-like contribution.

Finally, single crystal diffraction and polarised neutron diffraction [12] showed a very good atomic order in the samples, a very subtle change in the structure factors between 70 and 80 K, and allowed to determine the moments on Mn and Ni (3.8 and 0.2 μ_B respectively at 15 K) in agreement with the spin polarised band calculations [13,14]. Neutron inelastic scattering experiments were performed at Institut Laue Langevin, using 3 different crystals on the thermal beam IN8 triple-axis spectrometer and the high energy IN1 spectrometer [11]. A spin wave dispersion curve was followed up to about 100 meV. It begins with a quadratic dispersion law: $E = E_a + Dq^2$, with a very small anisotropy gap E_a (less than 1 meV). From neutron data, the stiffness constant D was obtained as 320 ± 20 meV Å^2 at 25 K, in good agreement with what can be deduced from the $T^{3/2}$ temperature dependence of the magnetisation (305 ± 40 meV Å^2) in the temperature range 4–70 K. Otto *et al.* [8] assumed a $T^{3/2}$ variation from 4 to 300 K and found a similar value: $D = 350 \pm 40$ meV Å^2. In addition to the spin wave dispersion curve, a diffuse intensity of magnetic origin was observed near the Brillouin zone boundaries for energies larger than 60 meV, which has been attributed to a continuum of Stoner excitations and will be analysed in the following sections.

3 New measurements

We measured the Hall effect, the magnetoresistance and the thermopower in NiMnSb in the low temperature range between 2 and 300 K, and reanalysed carefully a number of previous data in the low temperature range. The linewidth of ferromagnetic resonance was also measured on thin epitaxial films.

3.1 Hall effect

The Hall effect was measured by the AC current technique and lock-in detection under magnetic fields from -5 to 5 T. The normal Hall coefficient R_H^0 (Fig. 2) varies precisely as indicated by Otto *et al.* [9]. Its value between 120 and 300 K is nearly constant at about $+0.12 \times 10^{-10}$ ohm m/T, and rises up to $+0.75 \times 10^{-10}$ ohm m/T at 4 K. The positive sign indicates a dominant hole character. The anomalous Hall effect $\mu_0 R_H^a M_s$, where M_s is the spontaneous magnetisation increases with temperature from 80 to 300 K. This behaviour is in line with the observations of Otto *et al.*, who explained it in relation with the mechanisms of Hall effect [9], *i.e.* skew scattering and side jump phenomena. The amplitudes of these two contributions behave respectively as the resistivity and its square, explaining the increase of the anomalous Hall coefficient R_H^a with temperature from 100 K up to the Curie point. However, we observe an additional upturn of the anomalous term below 80 K (Fig. 2), which may correspond to the anomaly in the ratio of R_H^a/ρ reported by Otto *et al.* below 94 K. The increase in both Hall coefficients below 80 K should be attributed to a change in the electronic structure and/or to an increase in the mobility of carriers.

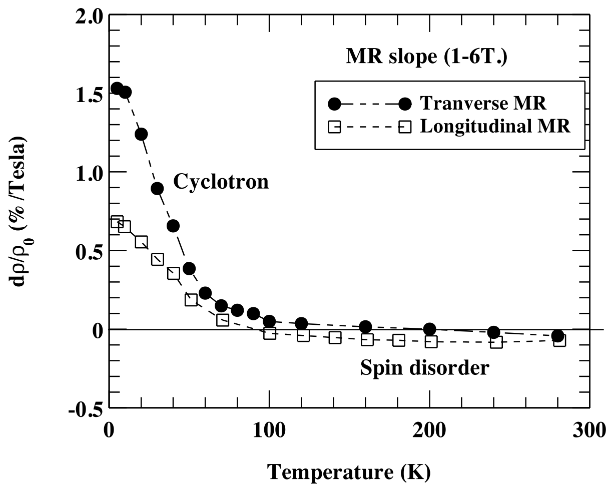


Fig. 3. Transverse and longitudinal magnetoresistances in NiMnSb (slopes of MR curves between 1 and 5 T).

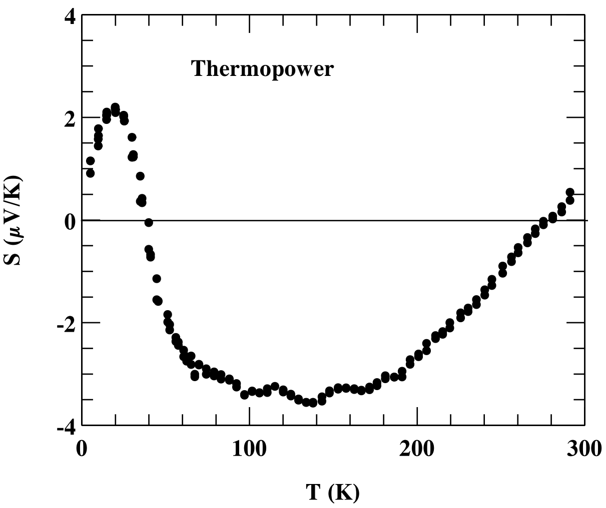


Fig. 4. Thermopower for NiMnSb.

3.2 Magnetoresistance

Longitudinal and transverse magnetoresistances (LG MR and TR MR) were measured under fields up to 5 T. After an initial weak drop of resistance in small fields (below 20 mT) which is probably related to the technical magnetisation, the low temperature magnetoresistance $MR = (\rho(H) - \rho(0))/\rho(0)$ is positive and a rather linear dependence on the field occurs between 1 and 5 T. At high temperatures, the MR turns negative. The slopes of MR curves between 1 and 5 T are reported in Figure 3 as function of temperature. We note that the transverse MR is higher than the longitudinal one; both drop rapidly from large positive values at low temperature to almost zero near 90 K, then there is a sign reversal and the LG MR turns negative above 100 K. These features indicate that cyclotron magnetoresistance is dominating at low temperatures. The TR magnetoresistance is known to be larger, at least for cubic polycrystalline samples, than the LG one, as the application of a transverse field continuously changes the mean drift velocity of carriers along the current axis. It is not possible to predict a ratio of the TR MR to LG MR, as the MR is strongly anisotropic for systems which, like NiMnSb, present open orbits on the Fermi surface [2]. The cyclotron MR is also expected to drop rapidly as the mean free path decreases, that is when the resistivity increases. However the amplitude drops more rapidly with temperature than would be predicted by a Kohler scaling, where the MR is a temperature independent function of $H/\rho(0)$, $\rho(0)$ being the zero field resistivity.

Conversely, the negative value of the MR indicates that the spin disorder contribution is dominating above 100 K; its amplitude should increase with temperature up to the Curie point.

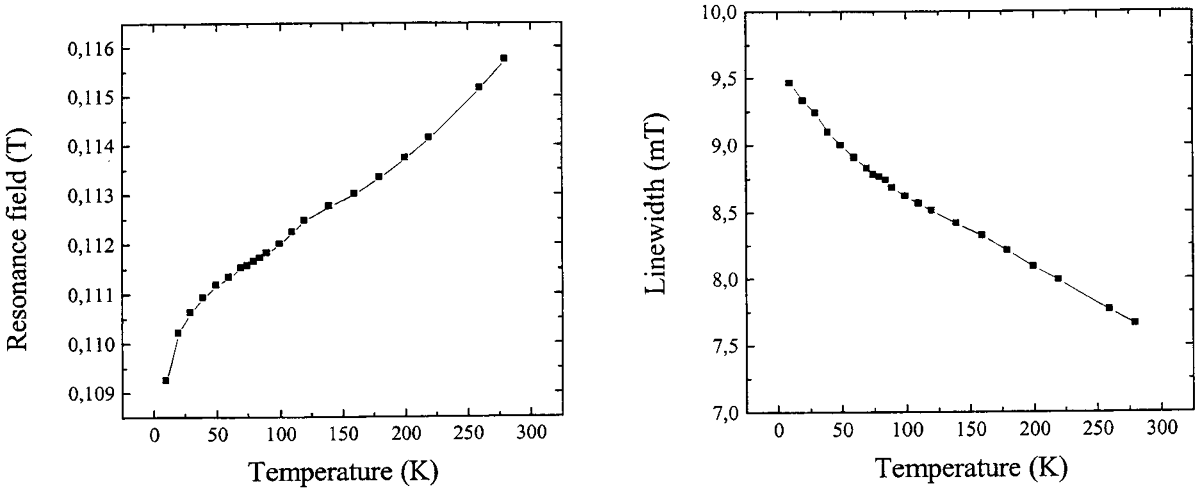
3.3 Thermopower

The thermopower of NiMnSb *versus* copper was measured by a direct comparison to that of a copper-constantan thermocouple. Its variations are given in Figure 4. The Seebeck coefficient gets more and more negative when lowering the temperature down to about 100–120 K, then it shows a rapid upturn and becomes positive below 40 K. Thus there is a change from electron-dominant to hole-dominant character as the temperature goes down. There is a disagreement with the sign of carriers deduced from the Hall effect data in the range 50–280 K; this comes most probably from the complexity of the real electronic structure which involves several bands, as we shall see.

3.4 Ferromagnetic resonance

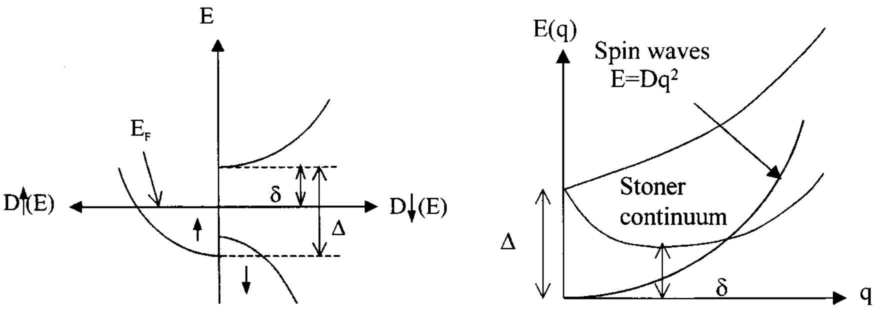
Epitaxial thin films, corresponding to the sequence NiMnSb(001)/Mo(001)/MgO (001), were grown using facing target sputtering at 400 °C. Details of the growth conditions and film structure can be found elsewhere [15]. A 500 Å thick film with lateral dimensions of 4×4 mm was used for this study. Ferromagnetic resonance data with the field perpendicular to the plane are consistent with the theoretical value of the demagnetising field. A weak in-plane anisotropy is observed, which may be related to the shape or magnetocrystalline anisotropy. The following data correspond to the case where the applied magnetic field lies in the plane of the film, parallel to the [100] axis.

The resonance field for $H \parallel [100]$ increases with temperature from about 0.109 T at 15 K to 0.116 T at 300 K (Fig. 5a), the rate of increase is much larger between 15 and 60 K than at higher temperature. Meanwhile the linewidth (Fig. 5b) decreases from 9.5 mT at 15 K to 7.6 mT. at room temperature; this decrease is rather uncommon even if we are dealing with a ferromagnetic system. The low value of the linewidth shows the very good homogeneity of the film. The resonance field for the field in the plane is given, neglecting the magnetocrystalline anisotropy, by: $(\omega/\gamma)^2 = H(H + M_s)$, where M_s is in tesla, ω is the resonance frequency, and $\gamma = 2\pi g\mu_B/h$ is the spectroscopic factor. When the spontaneous magnetisation M_s decreases from 0.95 T at 10 K, to 0.88 T



(a) (b)

Fig. 5. (a) Resonance field for $H \parallel [001]$ in the plane of a NiMnSb 500 Å thick film. (b) Linewidth for the NiMnSb film.



(a) (b)

Fig. 6. Schemes (a) for the band structure of half metallic ferromagnets, and (b) for magnetic excitations.

at 300 K, the theoretical resonance field should increase from 0.1034 T to 0.110 T, assuming $g = 2$. These values are not far from the experimental ones, thus the mean increase in resonance field can be reasonably accounted for by the variation of magnetisation. However, the more rapid variations of the resonance field and of the linewidth below 60 K must be related to some extra phenomenon, such as the anomalous variation of the magnetisation or some particular kind of relaxation mechanism. We shall develop this point in the discussion.

4 Discussion

4.1 Electronic structure

The half metallic character is due to the magnetic splitting Δ between up and down-spin subbands [1]. At low temperatures, the Fermi level for up-spins (majority spins) falls into a gap of the down-spin (minority) subband, as revealed by numerous band calculations which give the electronic structure at 0 K. Rising the temperature, the magnetic splitting Δ (about 2 eV at low temperatures) should decrease following the decrease in magnetisation. The energy difference δ between the Fermi level and the conduction band for down spins (Fig. 6a), which may be called the half-metal gap is a fraction of the gap for the down-spin semiconducting band. It will decrease faster than Δ, and the Fermi level will intersect the bottom of the down-spin conduction band at some temperature T^*, inducing a cross-over to a normal itinerant ferromagnet.

Most of the theory for itinerant magnets has been derived for weak ferromagnets [16], and only few theoretical studies have been devoted to the opposite case of strong (saturated Hubbard) ferromagnets. The relations between the magnetisation and density of states in NiMnSb have been worked out theoretically by Kulatov and Mazin [17], which predict that the compound should turn to a normal ferromagnet for a low decrease in magnetization. Irkhin and Katsnelson [18] examine the consequences of the spin splitting and the influence of spin wave excitations on the magnetisation, thermodynamic properties, relaxation rate of half metals. Otto *et al.* [19] treat these systems in a strong coupling model, they particularly state that "below some temperature T^*, spin flip scattering is not possible", and that "the onset of spin flip will lead to a rapid increase of the resistivity between T^* and the Curie point".

4.2 Magnetic excitations

Let us examine first the magnetic excitations in the low temperature half metallic phase. First transverse coherent spin wave excitations (magnons) occur, with a rather small energy gap E_a due to the anisotropy: according to neutron data, the gap is smaller than 1 meV (or 11.6 K). This gap should give a low temperature dependence of the magnetization as: $T^{3/2}\exp(-E_a/kT)$ instead of the $T^{3/2}$ classical spin wave law. However, no deviation from a $T^{3/2}$ law has been detected on the magnetisation curves along the 3 principal axes down to 2 K. It is also difficult to detect a $T^{4/3}$ correction to the magnetisation law, as calculated by Irkhin and Katsnelson [18], because this term would probably become important only at temperatures larger than the cross-over temperature to a normal ferromagnet. Let us note that a $T^{3/2}$ law has also been observed in the half-metallic CrO_2 compound between 1 K and 100 K [20].

Magnons are coherent intraband excitations, they can be described as excursions on the dispersion curves crossing the Fermi level, and at low temperature only low-q values (long wavelengths) are excited. Only weak relative deviations of the first neighbours moments occur, and locally all the spins including those of carriers remain in the up-direction, whereas the long range correlation function decreases. In the absence of anisotropy (Goldstone theorem), the magnetisation can rotate as a whole in any direction without energy barrier, the spin of current carriers follows and only a majority spin density still exists at the Fermi level. This rules out the apparent paradox between the occurrence of a vanishing down-spin density and that of spin excitations.

One magnon, corresponding to $\Delta S = -1$ for the whole lattice, scatters as well neutrons as current carriers, giving rise to a magnetic resistivity. As it was established by Mills and Lederer [21], the scattering of current carriers due to coherent spin waves in a ferromagnet varies as T^2 at low temperatures, which is in agreement with our observations. The T^2 behaviour is not restricted to this class of compounds, but also applies for instance to heavy fermions. In the half metal range, the resistivity channels corresponding to $\rho_\downarrow$ and $\rho_{\uparrow\downarrow}$ are negligible: $\rho_\downarrow$ corresponds to the resistivity of the semiconducting down-spin band, the gap of which may arise from charge transfer, hybridation or crystal field splitting, that is on binding energies. The $\rho_{\uparrow\downarrow}$ spin flip scattering corresponds to inelastic transitions across the gap δ which cannot be excited at low temperatures, but should only appear when kT becomes comparable to $\delta(T)$, that is just below T^*.

Besides spin wave scattering, there is also phonon scattering. In a temperature range well below the Debye temperature (estimated between 250 and 300 K), the phonon resistivity should behave as T^β, where the exponent depends on the nature of conduction electrons and on the shape of the Fermi surface. Generally $3 < \beta < 5$. The phonon contribution cannot be separated correctly from the magnetic one for NiMnSb, but in the isomorphous non magnetic compound NiTiSb the observed phonon exponent is 4 ± 0.2 for $T < 30$ K. The corresponding term becomes negligible at low temperatures compared to the observed T^2 one, which can be safely attributed to the spin wave contribution in the half-metallic system.

A second type of excitations, *i.e.* Stoner excitations, will arise at higher temperatures and superimpose a $\rho_{\uparrow\downarrow}$ contribution to the previous ones. They are interband and incoherent spin-flip excitations which have a more local character and form a continuum as a function of energy [22]. In normal ferromagnets, they occur at low temperature – and have been observed by neutron scattering down to less than 20 meV in the case of Fe [23] –, but their intensity is more significant at higher energies. In the case of half metals, their minimum energy is the distance δ between the Fermi level and the bottom of the conduction band for down-spin, as defined before (Fig. 6b). This minimum energy of the Stoner continuum occurs for a wave vector defined by the q-difference between the Fermi wavevector(s) of the majority band(s) and the reciprocal lattice vector corresponding to the bottom of the minority band (the X point of the Brillouin zone in the case of NiMnSb). For NiMnSb, these excitations should appear for a q-vector which is at about half the Brillouin zone boundary, which fairly corresponds to the neutron observations [11]. From neutron data, the maximum intensity of Stoner excitations occurs for an energy of about 60 meV for $T = 300$ K, but no direct measurement of the gap δ could be made as a function of temperature, as their cross section becomes much weaker at low energies, and as an intense flat optical phonon branch prevents their observation below 30 meV.

4.3 Interpretation of the anomalies in the physical properties

We now pretend that a cross-over from half-metal to normal ferromagnet occurs near 80 K. This will be justified by the variations in regime of the magnetisation and different galvanomagnetic properties.

First we can exclude, from the single crystal study, the fact that the various effects occurring near 80 K are due to a crystallographic change, or to a change in the magnetisation easy axis. The magnetisation regime above 80 K is that of itinerant magnets subject to spin fluctuations, and indicates a non-vanishing density of down-spin states. A similar explanation arises for the resistivity. Ueda and Moriya [24] have shown that the resistivity for spin fluctuation magnets varies as $T^{5/3}$ in an intermediate temperature range. This exponent is smaller than for the spin wave contribution, and is slightly larger than the one found experimentally (about 1.35). Note that in this range of temperature, the phonon contribution is much more significant than in the low temperature range and varies quite linearly with T, thus the observed exponent should be intermediate between those for the phonon, spin wave and spin fluctuation contributions.

It is interesting to note that the magnetisation is only slightly reduced at the transition temperature, being about 99.5% of $M_s(0)$. This reduction is the consequence of spin wave excitations only, it shows that the half metallic state in NiMnSb is extremely sensitive to the quasi-integer value of the magnetisation. The mean splitting Δ between up-and down-spin subband should vary in nearly the same proportions as the magnetisation, being thus slightly reduced at 80 K. However, the gap δ which is a small fraction of this splitting may vanish quite rapidly with the decrease of magnetisation.

The magnetoresistance turns negative near 100 K, when the spin flip scattering channel is opened, and the extrapolated residual resistivity A_2 in the high temperature range is higher than the value ρ_0 in the low temperature range due to this extra scattering. Finally, we note that both the thermopower and Hall effect turn to positive (or more positive) values when the temperature decreases: the minority electron pocket around the X-point in the Brillouin zone is shifted above the Fermi level and becomes unoccupied, thus reducing the electron character in the transport properties. Also the mobility of carriers should increase, as there is no more spin-flip scattering, which may explain the upturns observed for the Hall effect. However, there are still three bands of majority spin crossing the Fermi level [2], one of electron character, the two others correspond to open surfaces. This prevents any precise calculations of an effective density of carriers or mobility from the Hall effect and resistivity data, as previously recognised by Otto *et al.*

Concerning now the ferromagnetic resonance data, we observed anomalies on the temperature dependences for the resonance field and the linewidth. The peculiar variation of H_{res} can be partly explained by the more rapid variation of M_s below the cross-over temperature. A puzzling point is the temperature dependence of the linewidth, which also presents a more rapid variation in the half-metallic range. Lofland *et al.* [25] have studied the FMR in some magnetoresistive manganites. They observe in the case of $La_{0.67}Ba_{0.33}MnO_3$ annealed films a rather small linewidth, which slightly decreases with temperature as in the present case. These manganites are known to be highly polarised, or even half-metallic, and may behave in a similar manner concerning the linewidth.

The relaxation may be due to several causes: the Landau-Lifshitz-Gilbert relaxation, the spin-lattice relaxation, the spin-spin relaxation between local moments and conduction electrons (Korringa term), the exchange-conductivity effects. The first term gives a peak-to-peak width $\Gamma_{LLG} = 2/\sqrt{3}\omega G/\gamma^2 M$, where G is the Gilbert damping parameter [26]; this contribution does not vary in the right direction, as it should increase with decreasing magnetisation. The second term is generally taken as negligible in metals at low temperatures and should also increase with temperature.

Irkin and Katsnelson [18] have shown that the Korringa contribution to the longitudinal nuclear relaxation rate: $1/T_1 \approx T n_\uparrow(E_F) n_\downarrow(E_F)$ should be absent in a half-metal, and that the relaxation should be due to two-magnon processes which give a $T^{5/2}$ contribution. The Korringa relaxation is one of the most important contribution to the linewidth in metals; its absence should also lead to a reduction of the linewidth in electron resonance experiments. Indeed the linewidth is particularly narrow for the present metallic system. Nevertheless, the third mechanism should also lead to an increase of the linewidth with increasing temperature.

The fourth term can be expressed as $\Gamma = C(D\omega\sigma)^{1/2}$, where D is the spin wave stiffness and σ the conductivity. This term can be another important factor, as D and σ are quite large; it goes in the right direction as the resistivity increases with temperature, and with a larger rate below the cross-over temperature. Finally, the effects of some kind of bottleneck may be of importance, but the influence of such effects has not yet been investigated for half-metals

5 Conclusions

A coherent explanation of the anomalies encountered in various physical properties of NiMnSb near 80 K has been given in terms of a cross-over from a half metallic state to a normal metallic ferromagnet. Other experiments should be performed to complement these data, such as NMR experiments which so far have been performed only at rather high temperatures [27]. Efforts have been recently undertaken in order to apply half metals, including some ferromagnetic manganites, to spin electronics devices [4,5,28,29]. One of the shortcomings of such experiments is the loss of a full polarisation of electrons at high temperatures, which reduces the spin dependent scattering -or tunneling- amplitude. Besides problems related to the modification of the band structure at the interfaces, the cross-over to a normal metal at rather low temperatures may explain these facts. Despite a magnetisation value which is not much reduced compared to the saturation value (92% in NiMnSb at room temperature), the polarisation at the Fermi level may be considerably lowered once the minority spin subband becomes populated. One solution may be to find a compound where, on one hand, the Curie temperature is high (large Δ), on the other hand, the Fermi level falls at the bottom of the gap for the minority band (large δ). In this case, the half metallic character would perhaps last up to a higher temperature compared to the Curie point. A weak upturn on the Hall effect in PtMnSb below 100 K [9] seems to show that the half metallic character may also disappear at low temperatures in this similar Heusler compound, and thus the situation should not be better than for NiMnSb concerning room temperature applications. Perhaps a different behaviour may be expected in other types of half metallic systems, which undergo a metal-insulator transition at the Curie point [30,31].

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