Spin-dependent Peltier effect of perpendicular currents in multilayered nanowires

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Heat and charge transport perpendicular to Co/Cu multilayers are characterized by magnetoresistance and magnetothermoelectrical power. Furthermore, a very large voltage response to temperature oscillations under a dc current is observed, which depends strongly on the applied magnetic field. This effect is ascribed to a Peltier effect and its field dependence to a spin dependence of the Peltier coefficient.

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I. INTRODUCTION: THE INTERFACE TRANSPORT

The study of giant magnetoresistance[1,2] (GMR) with currents perpendicular to the planes[3] (CPP) has offered key data that established the pertinence of models involving spin-dependent conductivities.[4] It was shown earlier that the interaction of a spin-polarized current with magnetization affects also the thermoelectric properties of bulk magnets.[5,6] The thermodynamics of spin and heat transport through interfaces has been addressed more recently.[7] However, experimental data on the magnetic field dependence of the thermoelectric power of magnetic multilayers remain extremely scarce.[8]

In this paper, we investigate the spin-dependent heat and charge transport in magnetic multilayered nanowires by probing the thermoelectrical response to a flow of electrons crossing interfaces. That is, we measured the voltage V_{ac} caused by an alternating temperature, at zero alternating electric current, while a strong steady electric current I_{dc} forced the electrons through the multilayers. V_{ac} shows a linear and strong dependence on I_{dc}. This novel property of perpendicular transport is modeled with the standard formalism of the thermodynamics of out-of-equilibrium processes in the linear regime.[9] This analysis reveals that the observed slope $\partial V_{\mathrm{ac}}/\partial I_{\mathrm{dc}}$ results from the difference of the Seebeck coefficients of the two metals. Furthermore, we find that $\partial V_{\mathrm{ac}}/\partial I_{\mathrm{dc}}$ depends strongly on the magnetic field, that is, on whether successive magnetic layers are parallel or antiparallel. For the sake of clarity, we call this effect the magnetothermogalvanic voltage (MTGV). This observation points to the necessity of introducing spin-dependent Peltier coefficients, in a manner similar to the introduction of spin-dependent conductivities for the description of GMR.

II. EXPERIMENT

The samples are single nanowires, 6 μm long, with a diameter ranging from 30 to 60 nm, embedded in a polymer matrix. Each nanowire is composed of a stack of 300 bilayers of Co and Cu, 10 nm thick each, electrically connected at both ends to the macroscopic wiring via gold contacts. The synthesis process is detailed in Refs. 10–12.

The GMR is measured at a charge current of about 1 μA. The temperature of the wires under currents of high density was monitored by resistance measurements.[13] The temperature rise is of no more than a few K for wires about 50 nm in diameter.[14]

The magnetothermoelectrical power (MTEP) measurement shows the dependence on the magnetic field of the thermoelectric power, in other words, of the effective Seebeck coefficient of the multilayers. This measurement is performed by a lock-in amplifier detection carried out using a red laser light shining on one side of the membrane to produce a temperature gradient of a few K. The beam is chopped at 22 Hz, a frequency low enough to insure proper thermalization of the nanowire so that the results are frequency independent.[15] It induces an oscillation of the spatial average of the temperature of the nanowire with an amplitude T_{ac}, also of about a few K.

The thermogalvanic voltage (TGV) measures the ac voltage V_{ac} due to the temperature oscillation while a steady current I_{dc} runs through the nanowire. The dc current source insures that no ac current runs through the nanowire.

External magnetic fields are applied perpendicular ($\perp$) or parallel ($\parallel$) to the wire axis. All the magnetic responses presented below are calculated with the relation $[V_{H=0}-V(H)]/V_{H=0}$.

III. RESULTS

All the measurements presented in this study were performed at 300 K. Similar results were obtained at 15 K.[21] Figure 1 shows the data of sample A. The GMR ratio of about 15% at 300 K attests to the quality of the samples [Fig. 1(a)]. It can be accounted for with typical values of the asymmetry of the spin-dependent conductivities.[16] The MTEP ratio is about −20% [Fig. 1(b)]. This magnetic field dependence is accounted for either with an adaptation of the Mott formula[8] or by invoking a spin-dependent thermopower coefficient.[17]

The MTGV [Fig. 1(c)] adds a further challenge to the analysis. We note that the amplitude V_{ac}, linear with I_{dc},[21] is orders of magnitude stronger than the thermopower voltage when measured at −200 μA. One should keep in mind that V_{ac} is the response to the ac temperature oscillation. Hence, the temperature dependence of the resistivity $\partial R/\partial T$ contribute indirectly to the V_{ac} signal (see Sec. IV). However, as determined from independent measurements,[17,21] this contri-

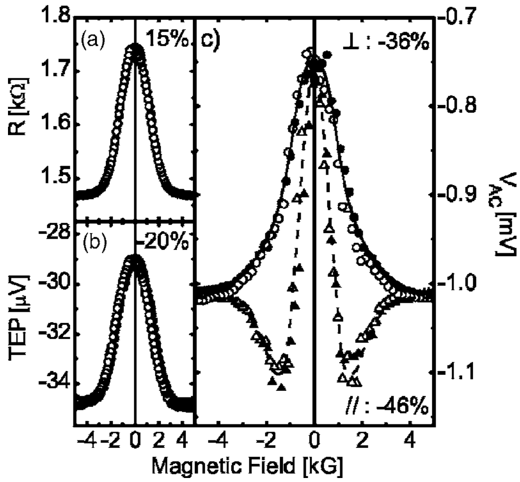

FIG. 1. Sample A. (a) GMR, (b) MTEP, and (c) MTGV curves at 300 K for magnetic field perpendicular (circles) and parallel (triangles) to the wire axis. Open (full) symbols refer to field sweep up (down). Lines are guides to the eye. The MTGV data are measured under a dc current of $-200\ \mu$A.

bution accounts for about $-300\ \mu$V in Fig. 1(c) [and about $-80\ \mu$V in Fig. 2(c)]. Therefore, most of the TGV signal has another origin than ohmic effects.

Measurements performed with perpendicular magnetic fields (in the plane of the layers) exhibit bell shaped curves, attributed to the progressive transition from the antiparallel (AP, at saturation field) to parallel (P, at zero field) configurations of the successive magnetic layers.

Unlike GMR and MTEP, the MTGV curves present a nonmonotonic field dependence when the magnetic field is applied parallel to the wire axis. This anisotropy is well illustrated in Fig. 2(c) for the sample D that exhibits a maximum MTGV response of about −85% in parallel fields.

Results for several samples are summarized in Table I. GMR and MTEP ratios are almost identical. MTGV ratios in

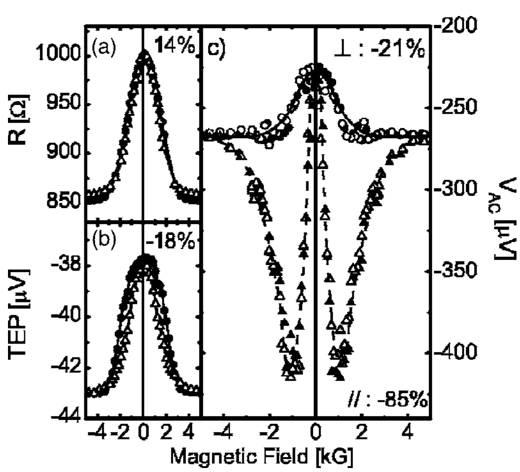

FIG. 2. Sample D. (a) GMR, (b) MTEP, and (c) MTGV curves at 300 K for magnetic field perpendicular (circles) and parallel (triangles) to the wire axis. Open (full) symbols refer to field sweep up (down). Lines are guides to the eye. The MTGV data are measured under a dc current of $-100\ \mu$A.

TABLE I. GMR, MTEP, and MTGV ratios at 300 K of various samples.

Sample	GMR	MTEP	MTGV (⊥)	MTGV (‖)
A	15%	−20%	−36%	−46%
B	14%	−17%	−25%	−44%
C	14%	−18%	−21%	−56%
D	14%	−18%	−18%	−85%

perpendicular magnetic fields are always larger. The large shifts of MTGV ratios observed between samples is attributed to the different contribution of the ohmic effects. It is also seen that MTGV ratios in parallel fields present a greater scatter.

These magnetic behaviors cannot be understood in terms of GMR since it was found that $\partial R/\partial T$ did not depend on the magnetic field. Hence, explaining the origin of this large voltage V_{ac} is our first challenge; understanding its magnetic field dependence is the second one.

IV. THE CPP-PELTIER EFFECT

The description of charge and heat currents by out-of-equilibrium thermodynamics in the linear approximation is well known. The constitutive relations can be inferred from a pure thermodynamic standpoint[9] or from a semiclassical description of conduction electrons in solids.[18] Following Ref. 9, we can write for the electric current density $j_e = I/A$, where I is the current and A is the cross section where it flows, and for the heat current density j_Q:

$$j_e = -\sigma \nabla V - \sigma\varepsilon \nabla T, \tag{1}$$

$$j_Q = \varepsilon T j_e - \kappa \nabla T \tag{2}$$

with σ and κ the electrical and thermal conductivities, respectively, and ε the Seebeck coefficient.

We consider now what these equations imply for a bilayer system composed of one ferromagnetic layer F and one nonmagnetic layer N. We impose an electrical current I through it. The Peltier effect arises at the junction of the two different metals from the mismatch of the Seebeck coefficients.

Contrary to Peltier coolers, in which the heat difference is normally compensated by external heat flux, we assume here that no heat exchange occurs with outside, i.e., that the interfaces are infinitely extended. Indeed, in a typical multilayer structure, the layers are disks of more than 50 nm in diameter and 10 nm in thickness. Furthermore, they are embedded in a polymer matrix and the interfacial heat resistance between metal and polymer is known to be quite large, whereas a good heat flux is insured along the metallic wire. Therefore we consider the heat fluxes to develop exclusively along the wire axis, perpendicular to the layers (CPP geometry) (Fig. 3).

In consequence, in the case of no externally imposed temperature gradient, we deduce from the continuity of the charge and heat currents across the interfaces that temperature gradients $\nabla T_{F,N}$ develop inside each layers as[21]

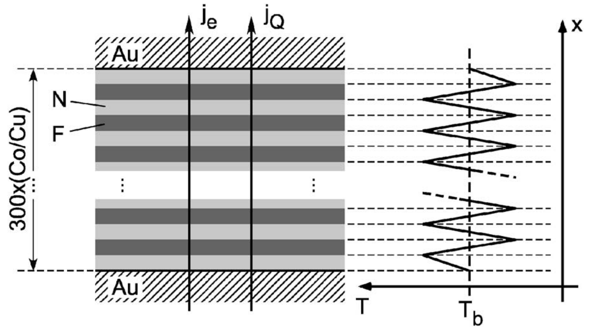

FIG. 3. Left: Co/Cu multilayer nanowire. The actual aspect ratio (stack height/diameter) of about 1000 insures charge (j_e) and heat (j_Q) currents perpendicular to the layers. Right: small temperature gradients induced by the Peltier effect [Eq. (3)].

$$\nabla T_F = \frac{\varepsilon_F - \varepsilon_N}{\kappa_F + \kappa_N} T \frac{I}{A} = - \nabla T_N. \tag{3}$$

Equation (3) expresses the combination of the Peltier effect $[(\varepsilon_F - \varepsilon_N)TI]$ and the heat conduction in each layer $(\kappa_F + \kappa_N)$. It implies that local temperature gradients develop in each layer, proportional to the charge current and opposite in sign in adjacent layers. The alternance of F and N layers gives the jigsaw temperature profile sketched in Fig. 3.

Likewise, the voltage drop across one bilayer can be calculated. Summing over all the bilayers of the nanowire, the overall voltage drop V along a nanowire of length L is

$$V = \frac{L}{2A}\left(\frac{1}{\sigma_N} + \frac{1}{\sigma_F}\right) I + \frac{L}{2A} \frac{(\varepsilon_F - \varepsilon_N)^2}{\kappa_F + \kappa_N} TI. \tag{4}$$

The first term is the overall resistance R. The second term is the sum of all the thermoelectrical powers induced by the local temperature gradients established in Eq. (3) that adds to the overall voltage drop. We call it the Peltier term since it derives directly from the Peltier effect.[22] For clarity, we chose to keep using the Seebeck coefficient ε instead of the Peltier coefficient $\Pi = \varepsilon T$.

This Peltier term is small and practically difficult to distinguish from the resistance. However, a lock-in detection enhances it. Through the temperature oscillation T_{ac}, the ac component of the voltage V_{ac} is derived from the relation $V_{ac} = T_{ac} \partial V / \partial T$. In the approximation of linear temperature dependence of the Seebeck coefficient $\varepsilon = (\delta\varepsilon / \delta T)T$, and under the condition $(\partial R / \partial T) T_{ac} \ll R$, verified in our samples, the slope (Fig. 2) is found to be

$$\frac{\partial V_{ac}}{\partial I_{dc}} = T_{ac}\left[\frac{\partial R}{\partial T} + \frac{L}{A\mathcal{L}} \frac{(\varepsilon_F - \varepsilon_N)^2}{\sigma_F + \sigma_N}\right]. \tag{5}$$

The thermal conductivity is written here in term of the electrical conductivity, according to the Wiedemann-Franz law $\kappa = \sigma \mathcal{L} T$, with $\mathcal{L}$ the Lorentz number. This simple approximation helps us in estimating the magnetic field dependence (Sec. V), but is not essential in identifying the origin of V_{ac}.

The first term in Eq. (5) takes into account the fact that the oscillation of the temperature of the wire implies a change of the resistance, thus a contribution to V_{ac} proportional to the derivative $\partial R / \partial T$. However, as mentioned in Sec. III, this ohmic contribution is limited. Hence, the main contribution to the TGV signal is the Peltier term, the second in Eq. (5).

V. THE SPIN-DEPENDENT CPP-PELTIER EFFECT

The observed MTGV is in essence the field dependence of $\partial V_{dc} / \partial I_{dc}$ [Eq. (5)]. Since $\partial R / \partial T$ is small, even negligible at low temperature,[21] and in any case independent of the magnetic field,[17,20] it does not contribute to the MTGV. Therefore the magnetic field dependence of the MTGV in Figs. 1(c) and 2(c) must be exclusively ascribed to the Peltier term.

In perpendicular magnetic fields, the only thing that changes between P and AP configurations is the spin of the conduction electrons relative to the magnetization of the layers. Thus, our MTGV measurements are detecting a spin-dependent Peltier effect. The dependence of MTGV on magnetic configurations can be understood with the two-current model, thereby assuming that the layers are thin compared to the spin diffusion length.[4,16] We define the spin asymmetry for charge (β) and heat (η) transport parameters as

$$\beta = \frac{\sigma_\downarrow - \sigma_\uparrow}{\sigma_\uparrow + \sigma_\downarrow}, \quad \eta = \frac{\varepsilon_\uparrow - \varepsilon_\downarrow}{\varepsilon_\uparrow + \varepsilon_\downarrow} \tag{6}$$

with $\sigma_{\uparrow(\downarrow)} = \sigma_0(1 \pm \beta)$ and $\varepsilon_{\uparrow(\downarrow)} = \varepsilon_0(1 \pm \eta)$. The classical combinations of the conductivities and the Seebeck coefficients for parallel and series currents[19] yield

$$\sigma_F^{P} = \frac{\sigma_\uparrow + \sigma_\downarrow}{2}, \quad \sigma_F^{AP} = \frac{2\sigma_\uparrow \sigma_\downarrow}{\sigma_\uparrow + \sigma_\downarrow},$$

$$\varepsilon_F^{P(AP)} = \frac{\varepsilon_\uparrow \sigma_{\uparrow(\downarrow)} + \varepsilon_\downarrow \sigma_{\downarrow(\uparrow)}}{\sigma_\uparrow + \sigma_\downarrow}. \tag{7}$$

The effective conductivities give the well-known GMR ratio of β^2. We derived from Eqs. (6) and (7) the MTEP and MTGV ratios, in the limit $\sigma_N \gg \sigma_F$ and $\varepsilon_F \gg \varepsilon_N$:

$$\text{MTEP} = \frac{-2\beta\eta}{1 - \beta\eta}, \quad \text{MTGV} = \frac{-4\beta\eta}{(1 - \beta\eta)^2}. \tag{8}$$

According to these relations, the GMR ratio of 15% yields a β of 0.44, which is reasonable.[16] From the MTEP ratio of −20% we deduce $\eta = -0.28$. These parameters imply a naturally higher MTGV ratio of −36%, which is consistent with our measurements. Therefore, at the two extreme configurations, P and AP, the amplitude of the MTGV is well understood with our CPP-Peltier model [Eq. (5)] and the spin asymmetries β and η deduced from separate measurements.

However, the evolution of the MTGV as the magnetization goes from the AP to the P configuration depends strongly on the orientation of the magnetic field [Fig. 1(c)]. This is in sharp contrast with the GMR and the MTEP that are well known to be isotropic. This difference indicates that MTGV detects a process that does not affect GMR and MTEP. When the field is parallel to the wires, and forces the magnetization out of the layers, one expects a more complex magnetization reversal than when the field is in the plane of the layers. It appears that MTGV detects this difference. A more elaborate model would be needed in order to account for this effect.

In summary, we investigated the thermoelectric properties of heat and charge transport in Co/Cu multilayer nanowires by means of MTGV experiments. The latter was demonstrated to be a local probe of the spin-dependent Peltier effects, in essence different from the GMR and MTEP. Connections between GMR, MTEP, and MTGV shifts between P and AP configurations are qualitatively described with the two-current model. However, the large MTGV ratios measured for fields parallel to the wire axis bring out spin dependent transport effects that are not observed in GMR and MTEP.

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[22] the Peltier term recalls the expression of the figure of merit $Z=\sigma\varepsilon^2 T/\kappa$ of thermoelectric devices.