Evidence for Weak Localization in the Thermal Conductivity of a Quasi-Two-Dimensional Electron System

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Evidence for weak-localization corrections to the electronic thermal conductivity of a quasi-two-dimensional electron system is reported for the first time. The validity of the Wiedemann-Franz relation theoretically predicted for such disordered systems is also experimentally verified.

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During the last decade, both experimental and theoretical investigations of the low-temperature electrical resistivity of weakly disordered electronic systems have led to quantum corrections to the Boltzmann formulation of electronic transport. It was found that these corrections become increasingly important as the amount of disorder increases, while the magnitude as well as the temperature dependence of the effect depend strongly on the dimensionality of the system. In fact, the corrections are larger in systems of lower dimensionality. For the particular case of quasi-two-dimensional (2D) systems, the resistivity increases logarithmically with decreasing temperature in the absence of spin-dependent processes. This nonclassical aspect of the carrier transport was theoretically interpreted in terms of two distinct quantum mechanisms: weak localization[1] and electron-electron interaction.[2] Soon after, expressions demonstrating the high sensitivity of weak localization to an external magnetic field were derived.[3]

Weak-localization and electron-electron-interaction effects in the 2D regime were initially observed experimentally in thin metal films,[4] electron inversion layers,[5] and semiconductor heterostructures.[6] The various aspects of the effect of disorder on the electron transport for different systems have been the object of recent comprehensive review papers.[7] In the last few years, such effects were evidenced in another important group of quasi-2D systems: bulk layered materials.[8-13] These materials exhibit highly anisotropic electronic transport properties. It was recently demonstrated that quasi-2D electron systems formed in low-stage acceptor graphite intercalation compounds[8-11] (GICs) and in pregraphitic carbons[12,13] are choice candidates to investigate disorder-induced quantum corrections to the electron transport.

Up to now, most experimental investigations on quasi-2D systems concerned the corrections to the electrical resistivity or to the Hall constant. As regards thermal transport, it was only very recently that theoretical estimates for the weak-localization and interaction corrections to the thermopower[14-16] and for the electronic thermal conductivity[17,18] for disordered systems were made. It was predicted that there were no quantum corrections to the Wiedemann-Franz law,

$$\kappa_E = L_0 T \sigma , \tag{1}$$

whatever the importance of the disorder, so that the electronic thermal conductivity κ_E should scale as the electrical conductivity at low temperature.[17,18] In relation (1), $L_0 = 2.45 \times 10^{-8}\ \mathrm{V^2 K^{-2}}$ is the free-electron Lorenz number and σ the electrical conductivity.

Generally speaking, the measured thermal conductivity κ is the sum of two contributions:

$$\kappa = \kappa_E + \kappa_L , \tag{2}$$

where κ_L is the lattice thermal conductivity.

Experimentally, the observation of the effects of localization and interaction on the electronic thermal conductivity of quasi-2D electron systems is a real challenge and has not been reported until now. Indeed, for semiconductor heterostructures heat conduction is expected to be dominanted by the lattice contribution κ_L. For disordered thin metal films, since there are no experimental data available, it is difficult to predict which of the two contributions, lattice or electronic, dominates. In any case, from the experimental side, whatever the intrinsic mechanism for heat conduction, the lattice contribution from the substrate of the thin films should thermally short circuit the sample. Thus, measurements of κ_E are prohibitively difficult to perform on these systems consisting of thin films.

On the other hand, layered materials could allow such extremely delicate experimental observations under favorable circumstances. Partially graphitic carbons are good candidates in this context. In these materials a mixture of two phases coexist. The first consists of well-stacked graphene planes in the *ABAB*... sequence characteristic of crystalline graphite with an interlayer spacing of 3.35 Å, while the second consists of randomly stacked graphene planes typical of a turbostratic structure.[19] In the latter case, the interlayer spacing is higher (3.44 Å) and the graphene planes in which conduction takes place are nearly uncorrelated. In recent works,[12,13] it was clearly shown that 2D weak localization occurs in

these materials and that the turbostratic phase is responsible for the 2D behavior as far as weak localization is concerned. Increasing the heat-treatment temperature of a partially graphitic sample above 2200 °C leads to a 2D-3D crossover due to a growing of the graphitic regions at the expense of the turbostratic regions.[13] In parallel, it is observed that the positive electrical magnetoconductance, which is about 20% in the liquid-helium range around 5 T for a nearly entirely turbostratic sample, progressively vanishes with increasing graphitic 3D ordering.[13] Magnetoconductance data have been convincingly interpreted in the framework of the weak-localization theory in the 2D regime over a wide range of temperature and magnetic field.[13]

Concerning heat transport, conduction in these materials is mainly governed by the lattice contribution which shows a steep temperature variation at low temperature. Consequently, it is almost impossible to observe directly the quantum correction to the thermal dependence of the electronic thermal conductivity. So, reliable information should come essentially from the transverse-magnetic-field dependence of the thermal conductivity at a given temperature, since the lattice thermal conductivity is not influenced by the external magnetic field.

Thermal conductivity measurements were performed using a static heat and sink four-probe method. Temperature gradients across the sample were generated by circulating an electrical current in a 10-kΩ metal-film resistance glued to a copper block in good thermal contact with the sample. At its other end, the sample was thermally bonded to the heat sink using GE 7031 varnish. The temperature gradient across the sample was measured using two carbon-glass resistors carefully matched to the sample by means of the GE varnish, allowing the determination of T_{cold} and T_{hot}, the temperatures of the cold and hot regions of the sample, respectively. The temperature of the heat sink was measured by means of a third carbon-glass resistor. The thermometers were first carefully calibrated in several separate runs to check their reproducibility upon cycling. The power dissipated in each carbon-glass sensor was limited to 10^{-9} W in order to avoid self-heating. In order to limit the heat losses by conduction through the electrical leads to less than 1%, 60-μm-diam Constantan wires were used to provide electrical connections to the heater and to the temperature sensors. At 3 K, the thermal conductance of the sample was found to be around 15×10^{-6} W/K. Electrical contacts to the sample for electrical conductivity and electrical magnetoconductance measurements were insured by means of Chromel wires and silver paste. The various voltages were measured using a Keithley K181 nanovoltmeter with a resolution of about 10^{-8} V.

As already pointed out by Sample, Brandt, and Rubin,[20] we found a rather complicated magnetoresistance behavior for the carbon-glass sensors. Indeed, we obtained positive magnetoresistance values for $T > 2$ K and negative values at lower temperature. Typically, at 4.2 K we obtained a magnetoresistance of 2.5% at 4 T, which corresponds to an apparent change in temperature of 3×10^{-2} K.

We determined the effect of a transverse magnetic field on the thermal conductivity of the sample in the following way. By pumping on the helium bath, the sample holder reached the desired temperature with a thermal stability of nearly 10^{-3} K. Then, we energized the heater to obtain a temperature gradient of about 0.5 K on the sample and we measured T_{hot} and T_{cold} at zero magnetic field and then at various magnetic fields. If

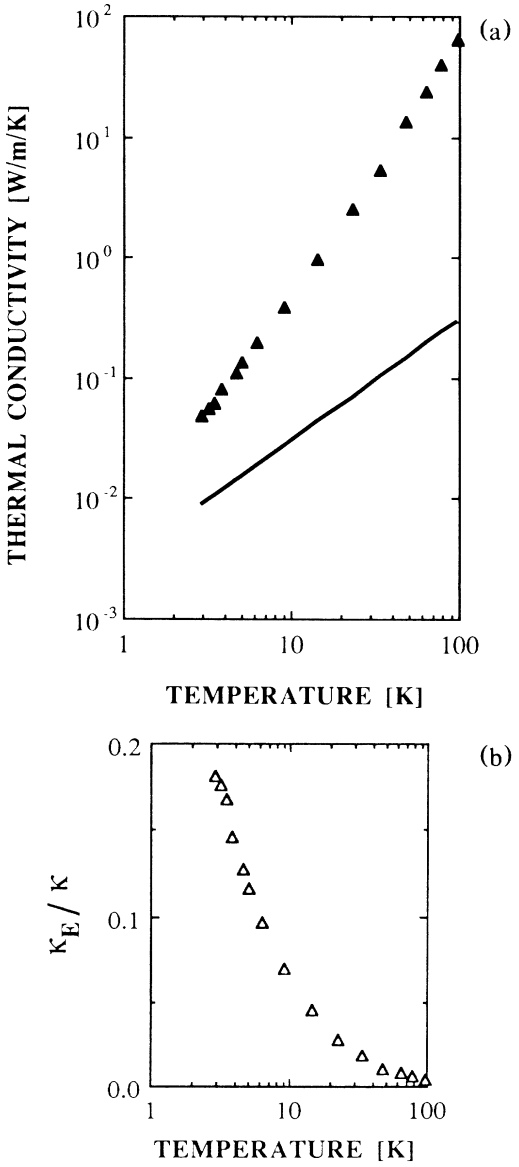

FIG. 1. (a) Temperature dependence of the measured thermal conductivity of a pregraphitic carbon sample heat treated at 1900 °C and of the electronic contribution to κ (line) calculated via relation (1). (b) Temperature variation of the ratio κ_E/κ.

necessary, small corrections to the current intensity applied to the heater were made to keep the power unchanged with increasing magnetic field up to 4 T. In the next step, we carefully determined the influence of the magnetic field on the sensors at T_{hot} and T_{cold}, respectively, for the same temperatures and fields at which measurements were taken, but with an isothermal sample, and applied the necessary corrections.

In Fig. 1(a) we present the temperature dependence of the as-measured total thermal conductivity κ in the temperature range $2 < T < 100$ K. The sample measured consisted of nearly fully turbostratic carbon heat treated at 1900 °C with a parallelepipedic geometry of $0.2 \times 5 \times 10$ mm^3.

In partially graphitic materials heat conduction is dominated by the lattice contribution from room temperature down to liquid-helium temperature.[21] Because of the extremely high Debye temperature of these materials ($\Theta_D \cong 2500$ K) and according to the microstructure of the sample, the lattice thermal conductivity reaches a maximum around room temperature with very high values (several hundreds of W m^{-1}K^{-1} according to the lattice perfection). At 4.2 K, instead, the lattice thermal conductivity may be smaller than 0.1 W m^{-1}K^{-1}, so that an observable electronic contribution to heat transport shows up. We see from Fig. 1(a) that, as expected,[21] the lattice thermal conductivity follows a T^n law with $n \sim 2$ from ~ 90 K down to ~ 8 K. In the liquid-helium temperature range, there is a departure from the T^2 law due to the contribution from the charge carriers.

We have also measured the temperature dependence of the electrical resistivity ρ of the sample in the same run. Then, assuming that the Wiedemann-Franz law [relation (1)] holds—an assumption that we shall discuss hereafter—we have calculated the electronic contribution to the thermal conductivity, κ_E, from the nearly temperature-independent electrical conductivity value ($\rho^{-1} \approx 1200\ \Omega^{-1}$cm^{-1}) measured at very low temperature. The calculated temperature dependence of κ_E is also reported in Fig. 1(a). In Fig. 1(b) we present the temperature variation of the ratio κ_E/κ for the same sample and temperature range as in Fig. 1(a). We see that the relative magnitude of κ_E is negligibly small around 100 K and increases with decreasing temperature to reach 18% around 3 K.

The electrical magnetoconductance at 4.2 K as a function of magnetic field up to 4 T is presented in Fig. 2. Previous works have shown that the electrical magnetoconductance of samples heat treated below 2200 °C is almost temperature independent in the liquid-helium temperature range due to the presence of magnetic impurities in the material.[12,13] The electrical magnetoconductance is positive over the whole magnetic-field range investigated and is nearly entirely due to the weak-localization effect. On the other hand, at zero field, the quantum fraction of the low-temperature electrical resistivity was estimated to be around 30% of the measured resistivity at low temperature.[12,13] So, by comparing thermal magnetoconductance data with electrical magnetoconductance data, the validity of the Wiedemann-Franz law for the disorder-induced quantum fraction of the resistivity may be directly checked. Since the electrical magnetoconductance reaches a value of 18% at 4 T and the calculated electronic contribution to the total measured thermal conductivity is also 18% at 3 K, we should expect a positive thermal magnetoconductance of a few percent below 4 T at this temperature. To be measurable, this very small effect requires a very careful calibration of highly sensitive thermometers, taking into account their sensitivity to the magnetic field, and the accurate determination of changes in thermal gradients of a few 10^{-3} K. This justifies the design of the special sample holder and the procedure described above, which are crucial for the observation of the effect.

In Fig. 2, we present the first evidence of a positive thermal magnetoconductance in the form of a field dependence below 4 T for a mean sample temperature of 2.9 K. Since for moderate magnetic fields the lattice is field insensitive, the thermal magnetoconductance should be entirely ascribed to the effect of the field on the electronic thermal conductivity. Assuming that the Wiedemann-Franz law [relation (1)] also holds in the presence of a magnetic field, as is generally the case, we may calculate the field dependence of the thermal magnetoconductance $[\kappa(H) - \kappa(0)]/\kappa(H)$ from that of the electrical magnetoconductance $[\sigma(H) - \sigma(0)]/\sigma(H)$ which is presented in Fig. 2. The result of this calcula-

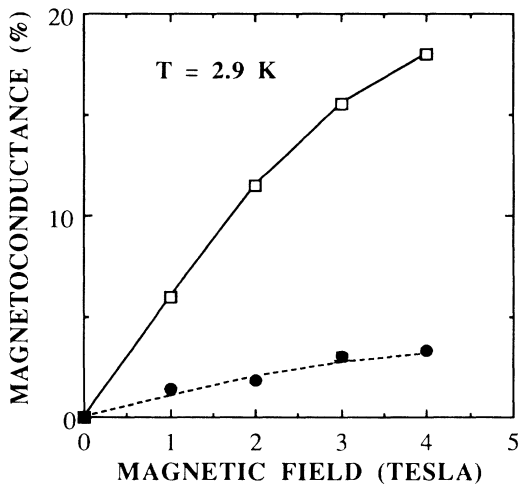

FIG. 2. Transverse-magnetic-field dependence (%) of the thermal magnetoconductance $[\kappa(H) - \kappa(0)]/\kappa(H)$ and of the electrical magnetoconductance $[\sigma(H) - \sigma(0)]/\sigma(H)$ at a temperature of 2.9 K. The open squares are the experimental electrical magnetoconductance data, while the solid circles are the measured thermal magnetoconductance data. The dashed line represents the thermal magnetoconductance calculated from the electrical conductivity results assuming the validity of the Wiedemann-Franz law (see text).

tion is represented by the dashed line in Fig. 2. If we compare the calculated values (dashed line) to the experimental data (solid circles), we find very good agreement within the experimental error ($\sim 0.3\%$).

The results show quite clearly that the underlying assumption, i.e., the validity of the Wiedemann-Franz law in the case of a 2D weakly disordered electronic system, is verified at low temperature. Thus, the quantum correction to the electronic thermal conductivity due to weak localization scales exactly as that to the electrical conductivity. In other words, it is experimentally evidenced that the Wiedemann-Franz law, which is always verified for the classical case in the elastic-scattering regime, applies also for weakly disordered systems, as theoretically predicted.[17,18] Indeed, it is found here that, within the 10% experimental uncertainty, a value of $L_0 = 2.45 \times 10^{-8}\ V^2K^{-2}$ is also obtained for the quantum correction to the conductivity. To demonstrate this, a very specific system has to be selected. It was found that the particular structure of disordered turbostratic carbons was a good choice, since it fulfills two essential conditions. It is a bulk material from which large samples can be cut, thus enabling thermal conductivity measurements to be performed, and it has an almost 2D electronic system presenting marked localization effects. Finally, as a corollary, we have also presented for the first time evidence for a positive thermal magnetoconductance in such systems.

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[1]E. Abrahams, P. W. Anderson, D. C. Licciardello, and T. V. Ramakrishnan, Phys. Rev. Lett. **42**, 673 (1979).

[2]B. L. Altshuler and A. G. Aronov, Solid State Commun. **30**, 115 (1979); B. L. Altshuler, A. G. Aronov, and P. A. Lee, Phys. Rev. Lett. **44**, 1288 (1980).

[3]S. Hikami, A. I. Larkin, and Y. Nagaoka, Prog. Theor. Phys. **63**, 707 (1980).

[4]G. J. Dolan and D. D. Osheroff, Phys. Rev. Lett. **43**, 721 (1979).

[5]D. J. Bishop, D. Tsui, and R. C. Dynes, Phys. Rev. Lett. **44**, 1153 (1980).

[6]B. J. Lin, M. A. Paalanen, A. C. Gossard, and D. C. Tsui, Phys. Rev. B **29**, 927 (1984).

[7]G. Bergmann, Phys. Rep. **107**, 1 (1984); P. A. Lee and T. V. Ramakrishnan, Rev. Mod. Phys. **57**, 287 (1985); B. L. Altshuler and A. G. Aronov, in *Electron-Electron Interaction in Disordered Systems,* edited by A. L. Efros and M. Pollak (North-Holland, Amsterdam, 1985), p. 1.

[8]L. Piraux, J. P. Issi, J. P. Michenaud, E. Mc Rae, and J. F. Marêché, Solid State Commun. **56**, 576 (1985); L. Piraux, V. Bayot, J. P. Michenaud, J. P. Issi, E. Mc Rae, and J. F. Marêché, Solid State Commun. **59**, 711 (1986).

[9]L. Piraux, V. Bayot, X. Gonze, J. P. Michenaud, and J. P. Issi, Phys. Rev. B **36**, 9045 (1987).

[10]L. Piraux, V. Bayot, J.-P. Issi, M. S. Dresselhaus, M. Endo, and T. Nakajima, Phys. Rev. B **41**, 4961 (1990).

[11]For a review, see L. Piraux, J. Mater. Res. **5**, 1285 (1990).

[12]V. Bayot, L. Piraux, J. P. Michenaud, and J. P. Issi, Phys. Rev. B **40**, 3514 (1989).

[13]V. Bayot, L. Piraux, J. P. Michenaud, J. P. Issi, M. Lelaurain, and A. Moore, Phys. Rev. B **41**, 11770 (1990).

[14]C. S. Ting, A. Houghton, and J. R. Senna, Phys. Rev. B **25**, 1439 (1982).

[15]J. W. P. Hsu, A. Kapitulnik, and M. Y. Reizer, Phys. Rev. B **40**, 7513 (1989).

[16]R. T. Syme and M. Pepper, J. Phys. Condens. Matter **1**, 2747 (1989).

[17]G. Strinati and C. Castellani, Phys. Rev. B **36**, 2270 (1987).

[18]M. J. Kearney and P. N. Butcher, J. Phys. C **21**, L265 (1988).

[19]M. S. Dresselhaus, G. Dresselhaus, K. Sugihara, I. L. Spain, and H. A. Goldberg, in *Graphite Fibers and Filaments,* Springer Series in Materials Science Vol. 5 (Springer-Verlag, Berlin, 1988), and references therein.

[20]H. H. Sample, B. L. Brandt, and L. G. Rubin, Rev. Sci. Instrum. **53**, 1129 (1982).

[21]C. A. Klein and M. G. Holland, Phys. Rev. **126**, A575 (1964).