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Electron coherence at low temperatures: The role of magnetic impurities

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Abstract

We review recent experimental progress on the saturation problem in metallic quantum wires. In particular, we address the influence of magnetic impurities on the electron phase coherence time. We also present new measurements of the phase coherence time in ultra-clean gold and silver wires and analyse the saturation of τ_{ϕ} in these samples, cognizant of the role of magnetic scattering. For the cleanest samples, Kondo temperatures below 1 mK and extremely small magnetic-impurity concentration levels of less than 0.08 ppm have to be assumed to attribute the observed saturation to the presence of magnetic impurities.

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1. Introduction

The understanding of the ground state of an electron gas at zero temperature is one of the major challenges in condensed matter physics. The conventional belief is that such a ground state is well described by Landau's theory of Fermi liquids [1]. This description of physical and electrical properties of a metal starts from the definition of quasiparticles, which are different from non-interacting electrons. These quasiparticles decay with a finite lifetime at a finite temperature. By definition, this lifetime has to diverge as temperature is lowered, being infinite at zero temperature.

Within the framework of Landau's theory of Fermi liquids, all scattering rates of an electron must vanish at zero temperature. As shown by Altshuler and coworkers [2], Landau's theory also holds in low dimensions and in presence of disorder.

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The phase coherence time is one of the fundamental concepts in mesoscopic physics. It is defined by the time an electron can travel in a solid before losing its phase coherence and thus its quantum, wave-like behaviour. Such decoherence is usually due to inelastic processes, like electron-phonon, electron-electron or electron-photon collisions. It has been shown by Altshuler and coworkers [2,3] that the phase coherence time of a weakly disordered quantum conductor diverges at zero temperature as electron-phonon, electron-electron and electron-photon interactions all go to zero at zero temperature. Therefore, the Fermi liquid description is also preserved for a disordered metal in low dimensions, precisely because of the diverging phase coherence times as temperature approaches absolute zero. Other dephasing mechanisms such as electron scattering from magnetic impurity spins or microwave radiation may generate a non-diverging or saturating temperature dependence of the phase coherence time.

This intuitive picture, however, has been challenged by experiments on metallic quantum wires which suggest that the phase coherence time τ_{ϕ} saturates at very low

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temperature [4]. Following this work, it has been argued that the observed saturation is indeed universal and intrinsic, and due to electron–electron interactions in the ground state of the Fermi liquid [5,6]. More recent studies of the phase coherence time in metallic silver quantum wires [7], on the other hand, show a relatively good agreement with the standard theory [2,3]. In these measurements, only small deviations of τ_{ϕ} compared with the standard theory have been observed at the lowest temperatures, which have been argued to be due to the presence of a very small amount of magnetic impurities.

In order to attribute the saturation of τ_{ϕ} in very clean metallic quantum wires to the presence of such a small amount of magnetic impurities, it is important to have an adequate theory which describes the underlying physics and enables a quantitative comparison with the experimental data. Quite recently, there has been tremendous progress in the understanding of the influence of magnetic impurities on the phase coherence time, both experimentally [8–11] and theoretically [12,13]. This new understanding raises the question: is it possible to reanalyse the temperature dependence of τ_{ϕ} in very clean metallic wires to determine whether the experimentally observed deviations from the standard theory can be explained within this picture of magnetic impurities.

The main purpose of this article is to review various experiments which have addressed the influence of magnetic scattering on electron coherence in metallic quantum wires. In the first part, we briefly review the pioneering experiments which studied electron coherence determined by magnetic scattering [14–17].

We then review recent measurements of electron coherence under high-magnetic fields ($\mu B \gg k_B T$) where the magnetic impurities are polarised and therefore should not contribute to dephasing [16–19].

In the last section, we present new data for the phase coherence time in ultra-clean gold and silver quantum wires, and compare them to presently existing data on equally clean samples from other groups. We then analyse the saturation observed in these samples assuming that the apparent saturation of τ_{ϕ} is due to magnetic impurities.

Our conclusion of this analysis is that, based on all presently available measurements of the phase coherence time in very clean metallic wires, it is hard to conceive that the apparent saturation of τ_{ϕ} is solely due to the presence of an extremely small amount of magnetic impurities.

2. Review of earlier experiments on magnetic impurities

As mentioned above, recent experiments invoke the presence of a small amount of magnetic impurities as a possible source of the frequently observed low-temperature saturation of the phase coherence time [7]. It is well known that the coupling of magnetic impurities to the conduction electrons gives rise to the well-known Kondo effect [20,21]. At temperatures above the Kondo temperature $T_{\rm K}$, the magnetic scattering due to Kondo impurities leads to a

very slow and an almost temperature-independent contribution to the dephasing time [22]. The magnetic contribution is maximal around the Kondo temperature [14,15] and decreases rapidly at lower temperatures [8,9]. Consequently, if a metallic sample contains a small amount of magnetic impurities with a very low Kondo temperature, the observed temperature dependence of τ_{ϕ} would show saturation at temperatures above $T_{\rm K}$.

Already in the early days of weak localization, many experimentalists have observed a systematic saturation of the electron phase coherence at low temperatures, when extracted from low-field magnetoresistance [23,24]. This saturation has often been attributed to the presence of some residual magnetic impurities [25], however, without any experimental verification.

To the best of our knowledge, the first experiment which clearly demonstrated the strong influence of magnetic impurities on electron coherence on the level of a few partsper-million (ppm) in very clean metallic samples has been carried out by Pannetier and coworkers in 1985 [26]. The need for very long phase coherence times in order to measure AAS oscillations in two-dimensional networks [27,28] pushed the authors to seek extremely clean metals to obtain very large values for the phase coherence length. The solution to the problem was to thermally anneal the samples. The annealing process oxidizes magnetic impurities, suppressing decoherence due to the Kondo effect, and therefore leads to an increase of the phase coherence length L_{ϕ} . The phase coherence length of two gold samples (before and after annealing) is shown in Fig. 1. One clearly sees the enhancement of the phase coherence length due to the annealing process [29].

These experiments therefore clearly show that the presence of an extremely small amount of magnetic

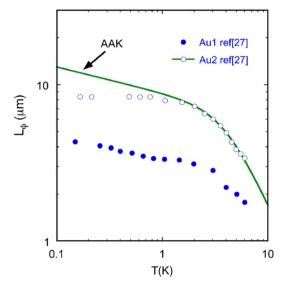


Fig. 1. Phase coherence length as a function of temperature for an ultrapure gold sample before (•) and after (o) annealing. The solid line corresponds to the theoretical expectation within the AAK picture [30]. Data are taken from Ref. [26].

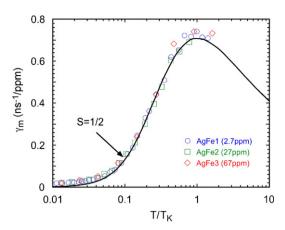


Fig. 2. Temperature dependence of the magnetic dephasing rate per impurity concentration $\gamma_{\rm m}$ of silver quantum wires doped with magnetic iron impurities of different ion concentrations. Data are taken from Ref. [10]. The solid line corresponds to the NRG calculation of τ_{ϕ} for the single channel, $S=\frac{1}{2}$ Kondo model [13].

impurities can lead to substantial electron decoherence at low temperatures, as expected. More importantly, even after the increase of the phase coherence length due to the annealing process, the phase coherence length still shows a relatively weak temperature dependence, much weaker than the theoretically expected temperature dependence due to electron–electron interaction [3].

A systematic study of the phase coherence time in the presence of magnetic impurities has been pioneered by Bergmann et al. [14] and van Haesendonck et al. [15]. Both groups have been able to measure the Kondo maximum of the dephasing rate due to the scattering of magnetic impurities in thin metallic films. Naturally, the measurements have then been extended to temperatures below $T_{\rm K}$ to investigate the ground state of the Kondo problem. In this limit $(T \leqslant T_K)$, Fermi liquid theory predicts a T^2 dependence of the inelastic scattering rate [31]. These experiments, however failed to observe this regime [32]. Instead, a very slow-temperature dependence of the magnetic contribution to the dephasing rate has been found below $T_{\rm K}$ [33]. This puzzle has remained unresolved for almost 20 years. In connection to the saturation problem [4], the questions of the Kondo ground state has recently gained new interest [8–13].

In a recent experimental study [8], a very curious linear temperature dependence of τ_{ϕ} below the Kondo temperature has been observed in AuFe quantum wires. An important contribution to the understanding of this behaviour came from theory, where an exact calculation of the inelastic scattering time in Kondo metals using Wilson's Numerical Renormalization Group (NRG) approach [12] was performed. The most interesting finding in these calculations was the fact that in a temperature range $0.1\,T_{\rm K} < T < T_{\rm K}$, the scattering rate due to magnetic impurities is linear in T, perfectly consistent with the experimental findings [8,9]. Only for temperatures below $0.1\,T_{\rm K}$, the T^2 temperature dependence is expected.

This temperature regime has been explored experimentally only very recently [10,11]. The experiment shows [10] that the dephasing rate due to the magnetic impurities is remarkably well described by the spin- $\frac{1}{2}$, single channel model [13] as shown in Fig. 2.

3. Experiments at high-magnetic fields

Another approach to investigate the influence of magnetic impurities on the electron coherence involves phase coherent measurements under strong magnetic fields. In this case, one can suppress the effect of magnetic impurities by applying a sufficiently high-magnetic field in order to fully polarise the magnetic impurity spins, and therefore study temperature dependence of phase coherence time with no contribution from magnetic impurity spins. Because of the required high fields in the order of teslas, low-field weak localization measurements cannot be used to extract the phase coherence time. However, measurements of Aharonov Bohm (AB) oscillations and universal conduction fluctuations (UCF) are possible. Pioneering work on both, UCF and AB oscillations in quasi-one dimensional (1D) quantum conductors containing a small amount of magnetic impurities (down to 40 ppm) has been performed by Benoît and coworkers in 1988 [16]. These data sets are shown in Fig. 3.

In this work, the authors clearly demonstrate that UCF as well as AB oscillations increase considerably at fields larger than 1 T, showing the suppression of the Kondo effect due to the polarization of the magnetic impurity spins. In the context of the present debate on the low-temperature saturation of τ_{ϕ} , these measurements have been repeated recently on metallic samples containing more dilute magnetic impurities [18].

In this experiment, the authors measure the magnetoconductance of copper rings and meander lines evaporated together. The phase coherence time extracted from weak localization measurements exhibit an anomalously strong saturation at low temperature for all samples investigated. The authors claim, however, that no evidence of the

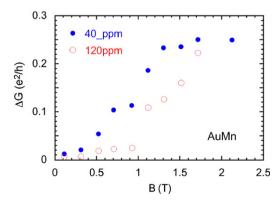


Fig. 3. Amplitude of the Aharonov–Bohm conductance oscillations versus magnetic field for two gold samples with different Mn impurity concentrations: 40 ppm (⋄) and 120 ppm (⋄). Data are taken from Ref. [17].

presence of Kondo impurities is detectable in the temperature dependence of the resistivity.

For the two samples showing the most pronounced saturation of the phase coherence time at low temperature, AB magnetoconductance oscillations have been measured on a ring structure at two different temperatures, $T\approx 40$ and $100\,\mathrm{mK}$. The magnetic field was swept up to $\approx 1.6\,\mathrm{T}$, i.e. a field sufficient enough such that $\mu_\mathrm{B}B\!\!>\!\!k_\mathrm{B}T$, with μ_B being the Bohr magneton. Superimposed on the UCF, AB magnetoconductance oscillations are clearly visible (see Fig. 3 of Ref. [18]). The amplitude of these oscillations clearly increases with magnetic field, as was observed by Benoît et al. [17] in their work on AuMn samples.

It is noteworthy that it is not straightforward to extract the phase coherence length directly from the amplitude of the AB oscillations, as the theoretical formula contains the phase coherence length both in a prefactor (as a ratio of $L_{\phi}/L_{\rm T}$, with $L_{\rm T}$ the thermal length) and in an exponential as the ratio of L_{ϕ} over the perimeter of the ring. Probably for this reason, the authors do not try to extract L_{ϕ} from their data but rather plot the amplitude of the AB oscillations as a function of the magnetic field and fit their curves using the formula:

$$\Delta G_{\rm AB} = C \frac{e^2}{h} \frac{L_{\rm T}}{\pi r} \sqrt{\frac{L_{\phi}}{\pi r}} \exp\left(-\frac{\pi r}{L_{\phi}}\right) \tag{1}$$

with C a dimensionless constant, e the charge of the electron, h the Planck constant, r the radius of the ring and $L_{\rm T}$ the thermal length, $L_{\rm T} = \sqrt{\hbar D/k_{\rm B}T}$ with D the diffusion constant and T the temperature. L_{ϕ} is obtained via the formula $1/\tau_{\phi} = 1/\tau_{\rm ee} + 1/\tau_{m}$, $\tau_{\rm ee}$ being the coherence time limited by the electron–electron interactions and τ_{m} the inelastic scattering time due to the presence of magnetic impurities. In the limit $(B, T \gg T_{k})$, the dephasing due to magnetic impurities is given by

$$\frac{\tau_m(B=0)}{\tau_m(B)} = \frac{g\mu_B B/k_B T}{\sinh(g\mu_B B/k_B T)}.$$
 (2)

There are obviously two unknown parameters in this problem, namely $\tau_m(B=0)$ and $\tau_{\rm ee}$. The authors assume that the saturation of the coherence time at low temperature is due to magnetic impurities; thus, they take $\tau_m(B=0) = \tau_\phi(B=0, T \to 0)$. As there is no direct experimental way to determine $\tau_{\rm ee}$, the authors take the theoretical value given by the AAK formula (Eq. (3); see Fig. 4). Using these values in Eqs. (2) and (1), they obtain a good agreement between their experimental measurements of $\Delta G_{\rm AB}(B)$ and the theoretical expression (1).

Note that, for the fitting procedure, the authors assume that the temperature dependence of the phase coherence time is given by the AAK formula for a quasi-1D wire:

$$\frac{1}{\tau_{\phi}} = \frac{1}{\tau_{\text{ee}}} + \frac{1}{\tau_{\text{ep}}} = AT^{2/3} + BT^{3},\tag{3}$$

where the second term corresponds to the electron phonon interaction. This temperature dependence of the $AT^{2/3}$ law, however, holds only for the case of quasi 1D wires. For the

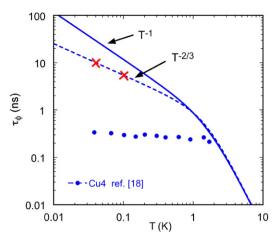


Fig. 4. Phase coherence time as a function of temperature for a copper wire extracted from weak localization measurements at low field and from AB oscillations at high fields. Data are taken from Ref. [18]. The dotted line corresponds to the AAK prediction for a quasi-one dimensional wire and the solid lines to the AAK prediction for a AB ring geometry in the regime $L_{\phi} < L$, where L is the perimeter of the ring [34,35]. The two data points (×) correspond to the theoretical values of the AAK theory for a quasi 1D wire, which have been used in Ref. [18] to fit the field dependence of the AB oscillations.

case of an AB ring, the correct temperature dependence for $1/\tau_{\rm ee}$ is in fact AT^1 (in the limit $L > L_{\phi}$) as pointed out recently [34,35]. If one plots the correct temperature dependence as shown by the solid line in Fig. 4, one clearly sees that the values for τ_{ϕ} used to analyse the highfield data (x in Fig. 4) lie much lower than the theoretical expectation. This experiment hence shows that the unusually strong dephasing observed in copper samples is definitely due to the presence of magnetic impurities [36]. However, it is clear that even under strong magnetic fields, the dephasing time does *not* recover the theoretical expectation within the Fermi liquid picture for the AB geometry [34,35]. In a strict sense, this experiment could even be interpreted as a support for additional dephasing at zero temperature apart from magnetic impurities: if under strong magnetic field, the "standard" description of dephasing due to electron-electron interactions developed by AAK is not recovered, there must be another mechanism which leads to decoherence at low temperatures [37].

Another experiment has been recently performed to clarify the role of magnetic impurities in the low-temperature saturation of the phase coherence time [19]. The fundamental difference with the previous experiment is that here the authors measure UCF in order to extract τ_{ϕ} and that they start from a "pure" sample: although the phase coherence time exhibits a clear saturation at low temperature, the resistivity as a function of the temperature follows nicely the $1/\sqrt{T}$ dependence predicted for the electron–electron interaction correction to the resistivity. For the UCF measurements, the authors use short and narrow wires in order to maximize the signal. Two different techniques are used to extract the phase coherence length: first, the phase coherence length can be directly related to

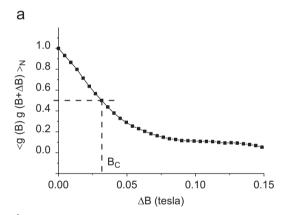
the amplitude of the UCF $\delta G_{\rm rms}$ via the relation:

$$L_{\phi} = \frac{3\pi}{8} \frac{L^3}{L_{\rm T}^2} \left(\frac{\delta G_{\rm rms}}{e^2/h} \right)^2, \tag{4}$$

where L is the length of the sample and $L_{\rm T}$ the thermal length. An alternative way involves the measurement of the correlation field $B_{\rm c}$ defined as the field over which the autocorrelation function $\langle G(B)G(B+\Delta B)\rangle$ reaches half of its zero field value. L_{ϕ} is then related to $B_{\rm c}$ via:

$$L_{\phi} = C \frac{h/e}{wB_{\rm c}},\tag{5}$$

where w is the width of the sample and C a numerical constant of order 1. In the experiment, the magnetic field is swept up to 15 T, i.e. $\mu_B B \gg k_B T$ as the temperature range was from 39 mK to 1 K. The phase coherence length extracted from the UCF using both technics (amplitude and correlation field) do exhibit a clear saturation of the phase coherence length at low temperature. From this experiment, the authors conclude that the saturation of the



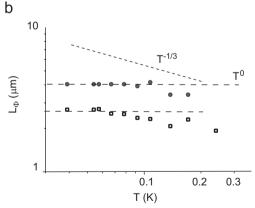


Fig. 5. (a) Normalized autocorrelation function calculated from conductance fluctuations above a field magnitude of 1 T. (b) Temperature dependence of L_{ϕ} for two samples, determined from the correlation field $B_{\rm c}$, shows saturation. The two quasi-one-dimensional gold wires have the following dimensions: 18 nm thick, 30 nm wide and 20 µm long, fabricated from gold with a purity of 99.9995%. The data shown in (a) is for a nanowire with a resistance of 2390 Ω . In (b), L_{ϕ} for this sample is represented by filled circles, while squares represent data for a second sample with a resistance of 2886 Ω . Diffusion constant for both samples is approximately $0.005\,{\rm m}^2/{\rm s}$. See Ref. [19] for additional details.

phase coherence time in their samples is not due to magnetic impurities but is rather intrinsic (Fig. 5).

What have we learned from this set of experiments (Benoît et al., Pierre et al. and Mohanty et al.)? From the first two, it is clear that magnetic impurities play an important role in dephasing, and that a strong magnetic field can suppress (or at least strongly reduce) such magnetic scattering. However, none of these experiments have been able to measure the temperature dependence of the phase coherence time under strong magnetic field. Moreover, following recent theoretical progress, one can state that in the AB oscillation [18] as well as in the UCF experiment [19] the phase coherence time determined at high fields is smaller than the one expected from the standard Fermi liquid picture. In addition, the experiment of Mohanty and Webb on pure wires seems to show that in their case the phase coherence time does saturate at lowtemperature even under strong magnetic field: one can thus conclude that, whether or not intrinsic, this saturation is not due to magnetic impurities. From this panorama, it seems that magnetic impurities do lead to strong dephasing and partially explain the low-temperature saturation of the phase coherence time, but these experiments do not provide evidences strong enough to rule out an intrinsic saturation as suggested by Mohanty and Webb in their pioneering work. Let us also mention that dephasing in much more disordered samples show also an anomalously strong dephasing [38] which is presently not understood.

4. Saturation of τ_{ϕ} in extremely pure metallic quantum wires

Let us now discuss the temperature dependence of τ_{ϕ} in extremely clean metallic quantum wires. In the absence of magnetic impurities, the main decoherence mechanisms in mesoscopic wires is assumed to be due to the electron–electron (ee) and electron–phonon (ep) interaction. The temperature dependence of $1/\tau_{\phi}$ for a quasi 1D diffusive wire is then given by the following expression [3]:

$$\frac{1}{\tau_{\phi}} = \frac{1}{\tau_{ee}} + \frac{1}{\tau_{ep}} = aT^{2/3} + bT^{3},$$

$$\frac{1}{\tau_{ee}} = a_{theo}T^{2/3}$$

$$= \left[\frac{\pi}{\sqrt{2}} \frac{R}{R_{K}} \frac{k_{B}}{\hbar} \frac{\sqrt{D}}{L}\right]^{2/3} T^{2/3},$$
(6)

The ep-term dominates at temperatures above 1 K whereas the ee-interaction term is the leading contribution at lower temperatures. Approaching zero temperature, the phase coherence time should hence diverge with a power law proportional to $T^{-2/3}$. On the other hand, almost all measurements on such wires show deviations from this commonly believed picture on electron dephasing. A few

 $\frac{1}{\tau_{\rm ep}} = bT^3.$

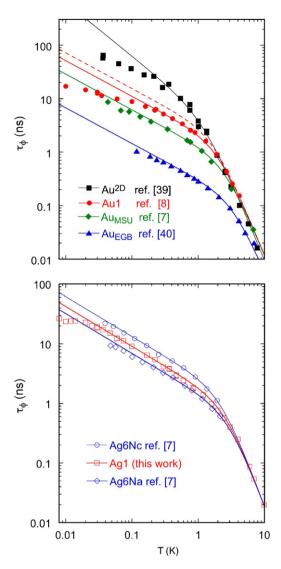


Fig. 6. Phase coherence time as a function of temperature for several metallic quantum wires. Top: gold wires, Bottom: silver wires. The solid lines correspond to the theoretical expectations within the AAK theory, using the experimentally determined parameters $a_{\rm exp}$ and $b_{\rm exp}$ given in Table 2. The dotted line corresponds to the AAK theory for sample Aul using the theoretically expected prefactor from Eq. (6).

examples from different experimental groups are shown in Fig. 6 and their electrical properties are summarized in Table 1.

In Fig. 6 we also plot the theoretical expectations, based on Eq. (6), as indicated by the solid lines. At sufficiently low temperatures, all samples show deviations from the theoretically expected prediction. Evaluating the theoretical parameter $a_{\rm theo}$, as listed in Table 2, we note that the experimental value $a_{\rm exp}$ determined from fitting the experimental data to Eq. (6) (solid lines in Fig. 6) is always lower than the expected theoretical value (dotted line). Stated differently, this means that the measured dephasing time is always lower than the theoretically expected value.

Naturally, the observed deviations have been associated with the presence of a tiny amount of magnetic impurities which contribute to dephasing at the lowest temperatures. As we have seen in Section 2, magnetic impurities lead to strong dephasing around the Kondo temperature due to inelastic magnetic scattering. If magnetic impurities with a sufficiently low Kondo temperature are for some unknown reason present in such a material, obviously one would obtain an apparent saturation of τ_{ϕ} in the investigated temperature range. To corroborate this picture, it is, however, necessary to have a quantitative analysis of such hypothetical Kondo impurities and to see whether such an assumption is reasonable.

Recent advances in the understanding of Kondo scattering in such systems [9,10,12,13] opens the possibility to reanalyse existing data on τ_{ϕ} in extremely clean metallic wires which show saturation and to verify whether this saturation can be explained by the presence of a tiny amount of magnetic impurities. As we have seen in Section 2, the temperature dependence of τ_{ϕ} in the presence of Kondo impurities is well described by the $S=\frac{1}{2}$ single channel Kondo model, at least down to temperatures of 0.1 $T_{\rm K}$ [10].

If magnetic impurities are somehow present in a metallic quantum wire, this can be taken into account in the temperature dependence of τ_{ϕ} by adding a third term in

Table 1 Sample characteristics: w, t, l, R, correspond to the width, thickness, length and the low-temperature electrical resistance, respectively

| Sample | w (nm) | t (nm) | L (μm) | $R(\Omega)$ | $D \text{ (cm}^2/\text{s)}$ | L_{ϕ}^{max} ($\mu\mathrm{m}$) | $\tau_{\phi}^{\mathrm{max}}$ (ns) | Ref. |
|------------------|--------|--------|---------|-------------|-----------------------------|---|-----------------------------------|-----------|
| Au ^{2D} | 22 μm | 22 | 44.3 mm | 3.980 | 135 | > 28.7 | >61 | [39] |
| Aul | 120 | 50 | 450 | 1218 | 241 | > 20.2 | >17 | [9] |
| Au_{MSU} | 90 | 45 | 175 | 1080 | 135 | >11.0 | >9 | [7] |
| Au_{EGB} | 90 | 15 | 100 | 3.0 | 135 | > 3.7 | >1 | [40] |
| Agl | 120 | 50 | 465 | 2997 | 105 | >15.8 | > 24 | This work |
| Ag6Na | 65 | 45 | 135 | 1440 | 115 | > 10.2 | >9 | [7] |
| Ag6Nc | 105 | 55 | 400 | 1440 | 185 | > 20.2 | > 22.0 | [7] |

D is the diffusion coefficient, L_{ϕ}^{\max} and τ_{ϕ}^{\max} the maximal values of the phase coherence length (phase coherence time). Note that the sample (Au^{2D}) [39] is two-dimensional and sample Au_{EGB} [40] consists of 21 narrow Au strips, connected in parallel. The resistance for this sample corresponds to the sheet resistance.

Table 2 Fitting parameters extracted from Eq. (6)

| Sample | $R(\Omega)$ | $D \text{ (cm}^2/\text{s)}$ | $a_{\text{theo}} \; (\text{ns}^{-1} \text{K}^{-2/3})$ | $a_{\rm exp}~({\rm ns}^{-1}~{\rm K}^{-2/3})$ | $b_{\rm exp} \; ({\rm ns}^{-1}{\rm K}^{-3})$ | |
|------------------|-------------|-----------------------------|--|--|--|--|
| Au ^{2D} | = | 135 | _ | 0.16 | 0.1 | |
| Au1 | 1218 | 241 | 0.28 | 0.38 | 0.08 | |
| Au_{MSU} | 1080 | 135 | 0.4 | 0.75 | 0.08 | |
| Au_{EGB} | | 135 | _ | 3.2 | 0.16 | |
| Ag1 | 2997 | 105 | 0.37 | 0.52 | 0.05 | |
| Ag6Na | 1440 | 115 | 0.55 | 0.67 | 0.05 | |
| Ag6Nc | 1440 | 185 | 0.31 | 0.35 | 0.05 | |

Note that the experimental values of a_{exp} differ slightly from the ones given in the original experiments. This comes from the fact that we fit all data such that a good agreement is obtained for the data at high temperatures ($T > 200 \,\text{mK}$).

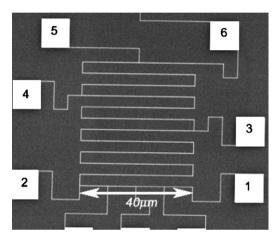


Fig. 7. SEM picture of a typical quantum wire used in our experiments. The current is fed into contact 1 and the voltage is measured at contacts 2 and 5. Contact 6 is set to ground.

Eq. (6). Eq. (6) reads then

$$\frac{1}{\tau_{\phi}} = \frac{1}{\tau_{\text{ee}}} + \frac{1}{\tau_{\text{ep}}} + \frac{1}{\tau_{m}}$$

$$= aT^{2/3} + bT^{3} + n_{\text{s}} * f(T/T_{\text{K}}), \tag{7}$$

where n_s is the impurity concentration in ppm and $f(T/T_{\rm K})$ the universal dephasing rate due to magnetic impurities, recently calculated by NRG [13]. As a and b are determined at relatively high temperatures ($T > 200 \, {\rm mK}$; see Fig. 6), the only adjustable parameters are the Kondo temperature $T_{\rm K}$ and the impurity concentration n_s . In the following, we will simulate the temperature dependence of the phase coherence time by assuming the presence of a small amount of magnetic impurities of concentration n_s and a Kondo temperature $T_{\rm K}$.

We will limit the analysis to our own data (sample Agl and Aul) which cover the largest temperature range. The same analysis could be done for all other existing data and one would arrive at the same conclusions.

A typical SEM picture of one of our samples is shown in Fig. 7. They are fabricated by standard electron beam lithography and lift-off technique. The metal (Au or Ag) is evaporated directly onto the Si wafer without adhesion layer. Special care has been taken for the sample design

such that there is no influence on the phase coherence due to the 2D contact pads [41]. The resistance is measured via a standard four-probe measurement technique using a lock-in amplifier in a bridge configuration. This is necessary as the resistance variations to be measured can be as small as $10^{-5}-10^{-6}$ of the absolute value. The resistance measurement of the sample is made in a current source mode: a voltage generated from a signal generator (typically at a frequency of 13 Hz) is fed into the sample (for instance, at contact 1) via a high resistance, typically of the order of $10-100 \,\mathrm{M}\Omega$. The voltage across the quantum wire is then measured between contacts 2 and 5 (contact 6 is at ground) and amplified by a home-made preamplifier (EPC1) situated at room temperature [42]. This voltage amplifier has an extremely low-noise voltage of about $0.4\,\mathrm{nV}/\sqrt{\mathrm{Hz}}$. We also take special care of the stability of the amplification stage. Very stable resistors (1 ppm/°C) are used for the current source. In order to avoid radiofrequency (RF) heating due to external noise, all measuring lines are extremely well filtered [43]. At the sample stage we have an attenuation of about -400 dB at 1 K. We have tested the efficiency of the attenuation by injecting a RF signal of frequency of 25 GHz with an amplitude of 25 dB into the sample cell. Weak localization measurements at 30 mK for sample Ag1, where there is still agreement with the AAK theory, have not been affected by such a procedure. All experiments have been performed in thermal equilibrium which means that the applied voltage across the *entire* wire is kept such that the inequality $eV \leq k_B T$ is satisfied at all temperatures.

In order to verify that the electrons of the samples are cooled down to the lowest measured temperatures, we measure the Altshuler–Aronov correction to the resistivity at very low temperatures [2,44]. As this correction is temperature dependent, it allows us to determine the effective electron temperature of our samples in situ. For the measurements, we apply a magnetic field of 40 mT in order to suppress weak localization correction to the resistivity.

In Fig. 8 we plot the resistance correction as a function of $1/\sqrt{T}$ for sample Au1. The resistance correction follows the expected $1/\sqrt{T}$ temperature dependence down to $10\,\mathrm{mK}$. Fitting the temperature dependence of the

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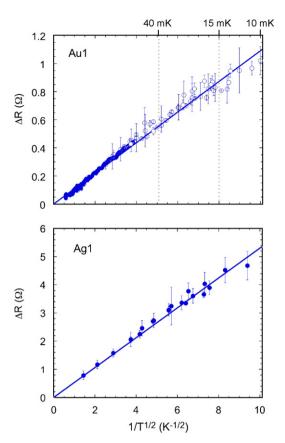


Fig. 8. Resistance variation as a function of $1/\sqrt{T}$ for sample Au1 (top) and sample Ag1 (bottom). The solid line corresponds to a fit using Eq. (9).

resistance correction to

$$\Delta R_{\rm ee}(T) = \alpha_{\rm exp} / \sqrt{T} \tag{8}$$

(solid line in Fig. 8), we determine α_{exp} and compare it to the predicted value [44] of

$$\Delta R_{\text{ee}}(T) = 0.782 \lambda_{\sigma} \frac{R^2}{R_{\text{K}}} \frac{L_{\text{T}}}{L}$$

$$= \lambda_{\sigma} \frac{\alpha_{\text{theo}}}{\sqrt{T}}, \qquad (9)$$

where $L_{\rm T} = \sqrt{\hbar D/k_{\rm B}T}$ is the thermal length and $R_{\rm K}=h/e^2$. The parameter λ_{σ} is a constant which represents the strength of the screening of the interactions in the metal under consideration. More precisely, it can be rewritten as a function of a parameter F which varies from 0 for an unscreened interaction to 1 for a perfectly screened interaction. In one dimension, one has $\lambda_{\sigma} = 4 - 3F/2$. The values of α_{exp} and α_{theo} are displayed in Table 3 [45]. From these values, we obtain $\lambda_{\sigma} \approx 3.18$ for silver and 2.62 for gold, giving a value $F \approx 0.6$ for silver in good agreement with theoretical values [46]. For the gold sample, however, we find $F \approx 0.9$, which is somewhat larger than expected. Note, however, that in many experiments the coefficient Fwhich can be deduced from the $R_{ee}(T)$ data [7] is obviously inconsistent with any theoretical model as it is often negative! A careful analysis of the available data is thus clearly needed in order to restore some coherence between

Table 3
Resistance correction to the resistivity due to electron–electron interactions

| Sample | R (Ω) | D (cm ² /s) | $lpha_{theo} \ (\Omega K^{-1/2})$ | $\begin{array}{c} \alpha_{exp} \\ (\Omega K^{-1/2}) \end{array}$ | λ_{σ} | n _s (ppm) | T _K (K) |
|--------|----------------|------------------------|-----------------------------------|--|--------------------|----------------------|--------------------|
| Au1 | 1218 | 241 | 0.042 | 0.11 | 2.62 | <0.08 | ≤0.01 |
| Ag1 | 2997 | 105 | 0.16 | 0.35 | 3.18 | <0.05 | ≤0.001 |

R is the electrical resistance and D is the diffusion coefficient. n_s (T_K) is the maximal ion concentration (maximal Kondo temperature) extracted from the fits shown in Figs. 10 and 11.

theory and experiment on this point of the e-e interaction correction to the resistivity in metals.

To determine the phase coherence time τ_{ϕ} we perform standard magnetoresistance measurements as a function of temperature. Typical measurements for sample Au1 and Ag1 are shown in Fig. 9.

For a quasi 1D wire the amplitude is essentially proportional to L_{ϕ}/L , where L is the length of the wire and L_{ϕ} is the phase coherence length. In addition, the width of the magnetoresistance curve corresponds to the situation where exactly one flux quantum h/e is introduced into a loop of a surface of $w \times L_{\phi}$ where w is the width of the wire and L_{ϕ} the phase coherence length. For the fitting of the magnetoresistance we use the following expression [44,47,48]:

$$\frac{\Delta R}{R} = \frac{2}{L} \frac{R}{h/e^2} \left\{ \frac{3/2}{\sqrt{\left[1/L_{\phi}^2 + 4/3L_{\text{so}}^2 + \frac{1}{3}(wBe/\hbar)^2\right]}} - \frac{1/2}{\sqrt{\left[1/L_{\phi}^2 + \frac{1}{3}(wBe/\hbar)^2\right]}} \right\},$$
(10)

where B is the magnetic field and $L_{\rm so}$ the spin-orbit length. In the low-temperature limit ($L_{\rm so} \ll L_{\phi}$), the effect of the spin-orbit scattering is to turn the weak localization into weak anti-localization (minimum in the magnetoresistance) [49,50]. We also note that the above formula is valid in the regime where $L_{\phi} < L$.

From fitting the magneto-resistance to Eq. (10) as indicated by the solid lines in Fig. 9, we can extract the phase coherence length L_{ϕ} . For the fitting procedure we first determine the spin–orbit length $L_{\rm so}$ at high temperatures, typically several kelvins where the spin–orbit length $L_{\rm so}$ is of the order of the phase coherence length (see inset of Fig. 9). For silver (gold) samples we obtain typically $L_{\rm so}\approx 500\,\mathrm{nm}$ (50 nm). We then fix the spin–orbit length and the only adjustable parameter for the fitting procedure is L_{ϕ} . The phase coherence time τ_{ϕ} is then obtained from the relation ($L_{\phi}=\sqrt{D\tau_{\phi}}$) and its temperature dependence for sample Au1 and Ag1 is plotted in Figs. 10 (top) and 11 (top). We also display the theoretical expectation based on the AAK theory as indicated by the solid lines. As clearly seen, for both data sets the experimentally measured value

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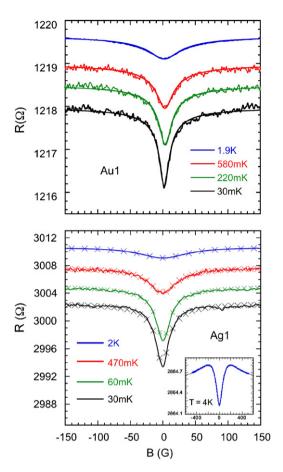


Fig. 9. Magnetoresistance of gold sample Au1 (top) and silver sample Ag1(bottom) as a function of temperature. The solid lines are fits to Eq. (10). As the fits are almost indistinguishable from the data points, we have highlighted the fits for sample Ag1 with symbols (×).

of τ_{ϕ} deviate substantially from the AAK prediction at the lowest temperatures. To see whether these deviations can be explained by the presence of a very small amount of magnetic impurities, we simulate the temperature dependence of τ_{ϕ} for the presence of a small amount of magnetic impurities by making use of Eq. (7). We first analyse sample Au1 which shows stronger deviations from the AAK theory. For the simulations we simply vary the possible ion concentration and the value of $T_{\rm K}$. The blue (a), red (b), green (c) and black (d) solid lines correspond to a simulation assuming $T_{\rm K}=40$, 10, 5 and 2 mK, with an impurity concentration of $n_{\rm s}=0.065$, 0.08, 0.1 and 0.18 ppm, respectively.

It is clear from our simulations that the temperature dependence of τ_{ϕ} can only be explained satisfactorily by the presence of magnetic impurities with a Kondo temperature $T_{\rm K} \leq 10 {\rm mK}$ and with a concentration smaller than 0.08 ppm. A possible magnetic impurity with a Kondo temperature in this temperature range is Mn ($T_{\rm K} \simeq 3 {\rm mK}$) [51] and one could hence assign the deviations at low temperatures to the presence of an ultra small amount of Mn impurities. The question is whether this assumption is reasonable and realistic to explain the observed deviations compared to the standard AAK picture?

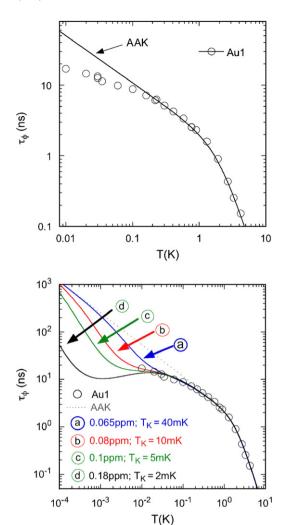


Fig. 10. Phase coherence time as a function of temperature for sample Au1 (\circ). The solid green line corresponds to the AAK prediction, the black (a), red (b) and blue (c) solid lines correspond to the NRG calculation assuming $T_{\rm K}=40,\,10,\,5$ and $2\,{\rm mK}$, respectively.

As the issue of low-temperature saturation is rather polemic and since the discussions are sometimes far from being scientific, we would like to take an objective stand to the saturation problem. In the following, we will give several comments to the above interpretation and point out several inconsistencies with this interpretation. The aim is not to give any definite answer to the dephasing problem, but to present experimental facts in a very objective way, and offer possible interpretations of the experimental findings.

If one assumes the presence of magnetic impurities with a Kondo temperature below the measurement temperature, this will lead to an almost temperature independent scattering rate for $T \geqslant T_{\rm K}$. Any experimentally observed saturation of τ_{ϕ} can therefore always be assigned to magnetic impurities with an unmeasurably low Kondo temperature. One could also argue, it is curious that for the

¹The exact temperature dependence is $\propto \ln^2(T/T_K)$ for $T \gg T_K$ and approaches a constant at T_K .

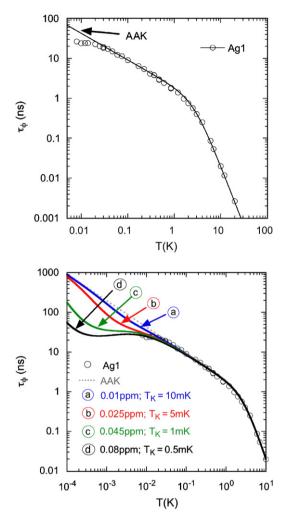


Fig. 11. Phase coherence time as a function of temperature for sample Ag1 (\circ). The solid green line corresponds to the AAK prediction, the black (a), red (b) and blue (c) solid lines correspond to the NRG calculation assuming $T_{\rm K} = 5$, 1, 0.5 and 0.1 mK, respectively.

case of gold wires, the observed temperature dependence of τ_{ϕ} can only be described satisfactorily by assuming the presence of one specific magnetic impurity with a Kondo temperature below the measuring temperature $T \leq 10\,\mathrm{mK}$, whereas it is known that the dominant magnetic impurity in gold is iron [52]. If we assume an additional iron concentration of the same order as, for instance Mn, the temperature dependence of τ_{ϕ} does not satisfactorily describe the experimental data [9].

The discrepancy actually gets worse if we analyse our results for the ultra-pure silver wire. This is displayed in Fig. 11. The green solid line corresponds again to the AAK prediction. At temperatures below 30 mK, our data deviate again significantly from the AAK prediction. The black (a), red (b) and blue (c) solid lines correspond to a simulation assuming $T_{\rm K}=10$, 5, 1 and 0.5 mK, with an impurity concentration of $n_{\rm s}=0.01$, 0.025, 0.045 ppm, and $c_{\rm imp}=0.08$ ppm, respectively. It is clear from our simulations that only magnetic impurities with a Kondo temperature *smaller* than 1 mK and with a concentration

smaller than 0.08 ppm describe satisfactorily the experimental data. This is indeed surprising, as the lowest known Kondo temperature for a Ag host is around 20–40 mK [53,72].

For the sake of completeness, we would like to point out that disorder introduces a modification for the spin relaxation rate at low temperatures. Ketteman and collaborators have suggested that magnetic impurities with very low Kondo temperature may be present in diffusive conductors since the Kondo temperature is strongly dependent on the local DOS [54]. Recent numerical simulations for 2D disordered metals suggest that the dispersion of Kondo temperatures induced by nonmagnetic disorder can enhance the spin relaxation rate at low temperatures compared to the case of a clean sample [73]. On the other hand, the distribution of T_K seems to be too small for the investigated samples. This scenario, however, requires a more systematic analysis of the distribution of Kondo temperatures in quasi 1D systems, in particular their dependence on the aspect ratio.

5. Comparison with theory

From what we have seen so far, it seems clear that magnetic impurities do play an important role in dephasing, and that this dephasing can be understood correctly within the framework of the Kondo physics. However, it also seems clear that an intrinsic saturation of the phase coherence time at low temperature cannot be ruled out from the sole experimental evidence available so far. Although controversial, a theoretical model has been developed by Golubev and Zaikin [5]. This model is an alternative to the standard AAK theory which claims that intrinsic decoherence at zero temperature is present even in a Fermi liquid type of framework. In this section, we would like to present the two models from a more formal point of view, stressing where the two approaches converge and where they diverge. However, we will not attempt here to compare these two models with the presented experiments. An interesting comparison of the theory of Golubev and Zaikin with experiments can be found i n Ref. [55].

In condensed matter physics, understanding the behaviour of an electron in its electromagnetic environment is an old problem. One of the central ideas is to integrate over the environmental degrees of freedom, thus encapsulating the effect of the environment on the particle into an influence functional. This method, originally due to Feynman [56], assumes that the environmental degrees of freedom are distinct from the electronic ones. This is obviously the case when they are of different nature (phonons, magnetic impurities). This is also the case when the test particle is indistinguishable from the environmental particles provided tunneling between the particle and the environment can be neglected as in the case of electrons of an external gate. A clear separation between system and environmental degrees of freedom is then restored and the

test particle only feels electromagnetic fluctuations created by charge and current density fluctuations in the gate.

In a pioneering effort, Guinea and coworkers have studied the influence of a normal Fermi liquid on charged particles [57], the dephasing of electrons in a diffusive wires under the influence of a nearby metallic gate [58] and the lifetime of electrons in ballistic quantum dots [59].

On the other hand, the theoretical debate triggered by experimental results of Mohanty et al. [4] concerns the coherence properties of an electron in a metal under the influence of electromagnetic fluctuations created by the other electrons. Because the test particle and the environmental electrons belong to the same electronic fluid, the standard influence functional approach is expected to break down since it does not incorporate exchange effects (Pauli principle). One has to deal with a fully interacting system of electrons coupled through the Coulomb interaction and possibly interaction with real photons.

Before turning to this difficult problem, let us recall that in 1D metallic systems, Coulomb interactions eventually lead to the breakdown of the Fermi liquid theory. Nevertheless, the effective theory describing the low-energy behaviour of the electronic fluid is well known: it is the Luttinger liquid theory [60]. Its stability under the presence of electric potential fluctuations induced by an external gate has been studied [61] and dissipation driven transitions towards two different phases have been predicted for sufficiently strong repulsive interactions. In the Luttinger liquid phase, decoherence induced by phonons or by real photons can indeed be computed exactly [62]. Therefore, the Luttinger liquid theory provides a very interesting theoretical framework for studying the behaviour of a dissipative strongly interacting electronic quantum system, showing how coupling to environmental degrees of freedom can change the fluid's properties or even trigger phase transitions and induce decay of Schrödinger cat states.

In the case of a 3D, 2D or quasi-1D diffusive conductor, the Fermi liquid theory is the only known low-energy effective theory for the electronic fluid. Consequently, the above questions are much more difficult to address than in the case of Luttinger liquid. This is exactly why raising the possibility of an intrinsic saturation of electron dephasing rate at very low temperature is so important: it would be a sign of the breakdown of the Fermi liquid picture which describes the ground state of metals, and associated low-energy properties. This obviously explains the intensity and the relevance of the theoretical debate raised by the work of Golubev and Zaikin [5,63].

In their work, Golubev and Zaikin (GZ) claim two results: (1) the correct derivation of an influence functional formalism for the dynamics of an electron in a diffusive conductor taking into account electromagnetic fluctuations created by the other electrons and the Pauli principle, and (2) the claim that these intrinsic electromagnetic quantum fluctuations lead to a saturation of the dephasing time of an electron in a diffusive conductor at zero temperature. After years of polemics, their derivation of an influence

functional taking into account the Pauli principle has been acknowledged first by Eriksen et al. [64] who used this tool to compute the renormalization of the effective mass for composite fermions and then by von Delft [65]. The GZ functional influence is an *important result* by itself since it potentially enables to account precisely the effects of quantum electromagnetic fluctuations on the electron dynamics whereas the original approach by AAK [3] only dealt with the effect of classical fluctuations (thermal Nyquist noise).

Because of these recent developments, the controversy has now been shifted to the second claim by Golubev and Zaikin since von Delft and his collaborators claim that the AAK results can be in fact recovered from the functional influence approach. They also argue that a simple Fermi Golden rule computation shows how the Pauli principle prevents intrinsic decoherence at vanishing temperature (see Section 5 of Ref. [66]). Some time ago, Golubev and Zaikin had already argued that their approach leads to terms that could not be obtained through the Fermi Golden rule approach and that those terms are precisely responsible for zero temperature dephasing [67]. But this criticism should not be confused with the point made by Montambaux and Akkermans [68] who showed that a proper treatment of the infrared cutoff in the Fermi Golden rule computation leads to a non-exponential phase relaxation of an electron in a quasi 1D disordered conductor treated within the AAK theory [3]. Also, more precise computations presented in Ref. [66] which take into account this effect also lead to the same non-exponential behaviour of decay functions for quasi 1D conductors. The real debate has to do with the effect of high-frequency environmental modes (with respect to k_BT/\hbar) within the influence functional approach. The basic argument against Golubev and Zaikin's results (2) is that, at vanishing temperature, an electron at energy ε above the Fermi surface cannot relax more than ε because of the Pauli principle. Thus, the Pauli principle provides an ultra-violet cutoff in the frequency integrals that determine decay functions and this forces the dephasing rate to vanish at zero temperature. Nevertheless, the von Delft et al. analysis has then been severely criticized by Golubev and Zaikin [69] who argue that von Delft and coworkers use a different influence functional which violates fundamentally important principles such as causality and detailed balance. At this point, it is not the purpose of the present paper to close this theoretical debate. Instead, we would like to clarify where the debate has been relocated and stress the difficulty of this theoretical problem.

First of all, from a theoretical perspective, the problem is far from being trivial. Because we are dealing with a many body interacting system, the GZ formalism is indeed a self-consistent approach to the electron dynamics: the influence functional itself depends on the properties of the electronic fluid and is basically derived in an operatorial form. It must be understood that evaluating and using this influence functional within a path integral formalism is nothing but

straightforward. The influence functional involves propagators with effective Hamiltonians containing an effective one-particle density operator which is expected to be non-local (in position). To deal with this problem, GZ rely on a phase space path integral. Eriksen et al. [64] have basically followed the same track and used the same method. On the other hand, von Delft [65] rely on a configuration space path integral (position only). In their comment [69], GZ point out that the resulting influence functional is different from theirs. In the same comment, they also mention several serious difficulties with the von Delft evaluation of the influence functional and thus seriously question the validity of von Delft's subsequent results. As mentioned before, resolving this controversy is beyond the scope of the present paper.

On the other hand, it is important to understand where the theoretical debate has shifted. Both Golubev and Zaikin, and von Delft and his collaborators agree on the existence of an influence functional which enables describing the many body problem within an effective one-particle formalism. They also agree on the definition of the influence functional under operatorial form. But their work diverge on the implementation of this object within a path integral formalism and the evaluation of the path integrals, needed to compute the weak localization correction to the conductivity.

Golubev and Zaikin rely on a phase space path integral which enables them to perform an explicit integration over the auxiliary fluctuating fields introduced by the Hubbard-Stratonovich transformation. A word of caution, phase space path integrals can be flawed when evaluating a propagator associated with an Hamiltonian whose kinetic term is not quadratic. As stressed before, the GZ influence functional precisely involves some effective Hamiltonians which are not quadratic in the particle's momentum. The well known and already less pathological example of a particle moving on a Riemannian manifold provides an illustration of these problems: the quantum mechanical amplitude is not given by the naive phase space path integral. Explicit evaluation of the phase space path integral requires an extra factor (reflecting the metric over path's space induced by the Riemannian metric) which usually cancels divergences arising in the naive phase space integral. Moreover, it requires going back to a discretized form which ultimately amounts to a choice of quantization [70].

On the other hand, von Delft uses the usual configuration space path integral which certainly provide a safer path with respect to the above-mentioned concerns. But the presence of a non-local operator in the effective Hamiltonians is a serious challenge to its explicit evaluation. Another way to avoid this complication by getting around the use of phase space path integrals for non-quadratic Hamiltonians has been proposed by GZ in Ref. [71] but it is not yet totally clear to us which of these lines of work provides a correct quantitative evaluation of intrinsic electron decoherence at very low temperature.

To summarize the theoretical debate suggested above, it seems reasonable to claim that the GZ influence functional defined in an operatorial way is correct and properly incorporates the effect of the Pauli principle as it has been demonstrated by von Delft. But, the precise use and evaluation of this influence functional within a path integral approach is a rather subtle question whose understanding will ultimately lead to a solution to the controversy over the discrepancy between GZ and AAK results.

6. Conclusion

In this article, we have reviewed recent experiments on dephasing in metallic quantum wires in regards to magnetic impurities. From the data we have presented, it is clear that magnetic impurities lead to strong dephasing at low temperatures. However, all experiments presented here strongly indicate that there must be an additional mechanism which leads to dephasing at low temperature apart from magnetic impurities. Let us therefore emphasize that at present there is no experimental evidence against zero temperature saturation. In our opinion, it does not make much sense to look for cleaner and cleaner materials to see whether there is a saturation of τ_{ϕ} at the lowest temperatures: indeed, the controversial theory by Golubev and Zaikin [5] predicts a D3 dependence of the zero temperature saturation time τ_0 [6], with D being the diffusion coefficient. Going to cleaner and cleaner materials would obviously increase the diffusion coefficient and the saturation, if existing, would never be observable at presently accessible temperatures. Instead one should try to look for a system where the diffusion coefficient can be varied over possibly one decade in T without changing the purity of the system. This was attempted in the original paper by Mohanty and Webb.

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