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## LETTER TO THE EDITOR

## Saturation of electron dephasing in three-dimensional polycrystalline disordered metals

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### Abstract

We have systematically investigated the low-temperature electron dephasing times  $\tau_\phi$  in more than 40 three-dimensional polycrystalline impure metals with distinct material characteristics. In all cases, a saturation of the dephasing time is observed below about a (few) degree(s) Kelvin, depending on samples. The value of the saturated dephasing time  $\tau_0 [\equiv \tau_\phi(T \rightarrow 0 \text{ K})]$  falls basically in the range 0.005 to 0.5 ns for all samples. Particularly, we find that  $\tau_0$  scales with the electron diffusion constant  $D$  as  $\tau_0 \sim D^{-\alpha}$ , with  $\alpha$  close to or slightly larger than 1, for over two decades of  $D$  from about 0.1 to 10 cm<sup>2</sup> s<sup>-1</sup>. Our observation implies an essentially constant saturated dephasing length of  $\sqrt{D\tau_0} \sim 1000 \text{ \AA}$  in three-dimensional polycrystalline disordered metals. A complete theoretical explanation is not yet available.

The issue concerning the saturation of conduction electron dephasing time  $\tau_\phi(T)$  in disordered conductors and mesoscopic systems has recently attracted renewed theoretical [1–4] and experimental [5–8] attention. In particular, it is of great interest whether or not the saturation of  $\tau_\phi$  might be universal in all material (e.g. polycrystalline metal, amorphous metal, and MBE-grown semiconductor) systems and in all (zero, one, two and three) dimensions. It is also important to ask whether or not there might be a common, intrinsic source that causes the (almost) universally observed saturation. The interest in this issue of the existence or not of a *finite* conduction electron dephasing time at absolute zero temperature is closely connected with the concern about the validity of the Fermi-liquid behaviour, the possibility of the occurrence of a quantum phase transition, and also the feasibility of quantum computing [9]. There are also works suggesting that this might explain the long-standing persistent current problem in metals [10]. In practice, the value of  $\tau_\phi$  can be reliably determined using quantum-interference studies [5–8] such as weak-localization effects and universal conductance fluctuations. It can also be determined using studies of the shape change of the electron distribution function upon applying a bias voltage across the conductor [11].

In this work, we have systematically investigated the electron dephasing times  $\tau_\phi$  in numerous *three-dimensional polycrystalline* impure metals. Our samples were made of various materials using various fabrication techniques at different laboratories. Also, the samples were

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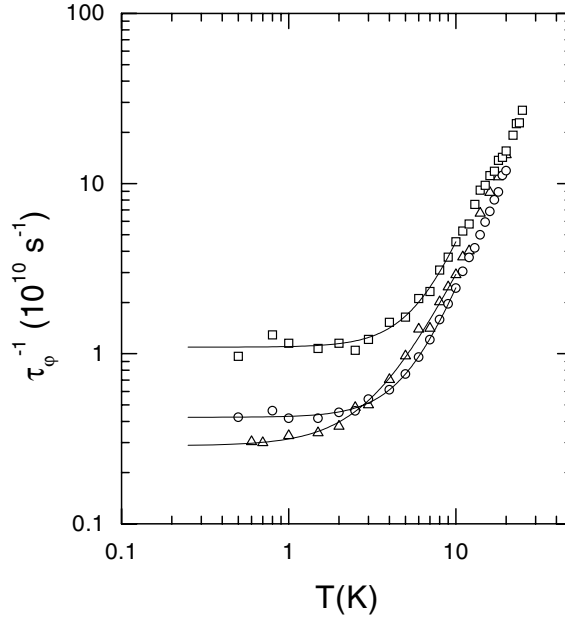
measured at different laboratories at very different times. Our samples considered in this work include dc sputtered thick  $\text{Au}_{50}\text{Pd}_{50}$  films, dc and/or RF sputtered thick  $\text{Ag}_{40}\text{Pd}_{60}$  films, dc sputtered thick Sb films, thermal-evaporation deposited thick  $\text{Au}_x\text{Al}$  films ( $1.8 \lesssim x \lesssim 2.2$ ), thermal-evaporation deposited thick  $\text{Sc}_{85}\text{Ag}_{15}$  films, and arc-melted  $\text{V}_{100-x}\text{Al}_x$  alloys ( $20 \lesssim x \lesssim 24$ ). The thick film samples are typically of the order  $4000 \text{ \AA} \times 0.3 \text{ mm} \times 17 \text{ mm}$ , while the arc-melted samples are typically on the order  $0.1 \times 0.1 \times 10 \text{ mm}^3$ . Since more than 40 samples are studied, it would not be practical to list the material parameters for all the samples. However, we notice that all of the samples studied are made from very high-purity starting materials obtained from reputable suppliers such as Alfa Aesar, Cerac, and Goodfellow. Since the major aim of this work is to study experimentally whether or not there might exist a universal behaviour of the saturated (or, the zero-temperature) dephasing time  $\tau_0 [\equiv \tau_\phi(T \rightarrow 0 \text{ K})]$ , the use of many kinds of samples with very distinct characteristics should therefore serve this purpose well. Any behaviour of  $\tau_0$  common for all these materials, if observed, must be a manifestation of the very general nature of the zero-temperature dephasing time. In addition, we notice two more practices regarding our experimental method. (a) Our measurements of  $\tau_\phi$  had been performed over a long time period of over four years (1997–2000). (b) Our measurements of  $\tau_\phi$  were carried out at two different laboratories located at two different sites 80 km apart. Nevertheless, the same  $^3\text{He}$  fridge and the same electronic measuring systems (except with different grounding and shielding) were used for all measurements. Surprisingly, regardless of the above-mentioned very different preparation and measurement conditions, it is noteworthy that our  $\tau_0$  measured in all samples varies with the electron diffusion constant  $D$  with a simple power law as  $\tau_0 \sim D^{-\alpha}$ , with  $\alpha$  close to or slightly larger than 1 (see below). Such universal behaviour could not be just ‘accidental’ and certainly deserves serious theoretical and experimental attention. We emphasize that we are concerned with three-dimensional polycrystalline metals. It is our opinion that the saturation behaviour (e.g. the functional form of  $\tau_0$  on disorder) is universal for a given dimensionality and a given kind of sample (e.g. polycrystalline or well-textured semiconductor) structure, while it might not be universal over different dimensionalities and different sample structures.

We have measured the low-field magnetoresistances at liquid-helium temperatures and compared them with three-dimensional weak-localization predictions to extract the values of  $\tau_\phi(T)$  for our samples. The details of the data analysis procedure has been discussed previously [12]. Here we merely stress that, for every sample studied in this work, the three-dimensional weak-localization predictions can well describe our measured magnetoresistances. Thus,  $\tau_\phi$  can be very reliably extracted. Figure 1 shows the total electron dephasing rate  $\tau_\phi^{-1}$  as a function of temperature for three representative thick  $\text{Ag}_{40}\text{Pd}_{60}$  films with resistivities as indicated in the caption to figure 1. The symbols are experimental data and the solid curves (emphasizing particularly the low measurement temperatures) are the least-squares fits to the form

$$\frac{1}{\tau_\phi(T)} = \frac{1}{\tau_0} + \frac{1}{\tau_i(T)} \quad (1)$$

where we assume that the total dephasing time comprises two terms, with  $\tau_0$  being a temperature-independent (i.e. the saturated) dephasing time which dominates at the lowest measurement temperatures, and  $\tau_i$  being the relevant inelastic scattering time which is usually dominant at a (few) degree(s) Kelvin or so and higher, depending strongly on samples. One sees that, for every sample,  $\tau_\phi^{-1}$  first decreases with decreasing temperature in the high-temperature part of our measurements. This decrease in the total dephasing rate can readily be ascribed to the weakening of the inelastic scattering process with decreasing temperature. That is,  $\tau_\phi^{-1}$  is dominated by  $\tau_i^{-1}$  and it decreases with weakening  $\tau_i^{-1}$  above 3 K or so for these particular samples. At lower temperatures, the inelastic scattering becomes weaker and weaker so that

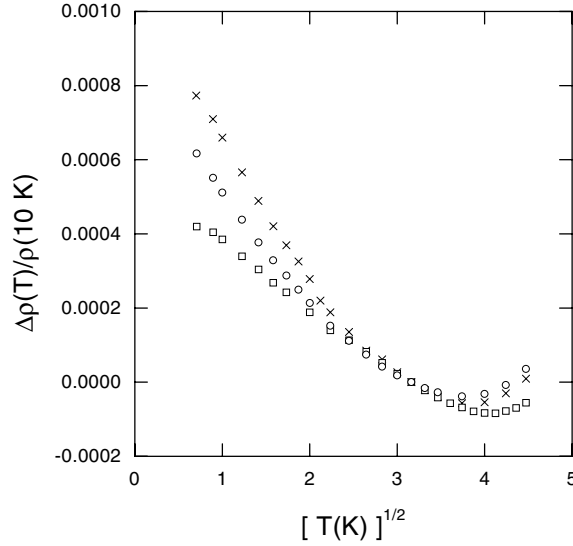
$\tau_\phi^{-1}$  becomes more and more dominated by the temperature-independent  $\tau_0^{-1}$ . Inspection of figure 1 reveals that equation (1) can well describe the experimental data. In fact, in some cases (e.g. figure 1) when the saturation of  $\tau_\phi$  already sets in at a few degrees Kelvin high,  $\tau_0$  can already be determined using essentially the measured value at the lowest temperatures. This ‘advantage’ of relatively *high*-temperature saturation largely reduces the uncertainties in the least-squares fits, making the inferred values of the adjusting parameters  $\tau_0$  and  $\tau_i$  very reliable. (Another great advantage of the present experiment is that we have intended to use many samples possessing strong spin-orbit scattering. In the limit of strong spin-orbit scattering, it is known that  $\tau_\phi(T)$  is the *only* adjusting parameter in the comparison, i.e. least-squares fits, of the measured low-field magnetoresistances with the weak-localization theoretical predictions, making the extraction of  $\tau_\phi(T)$  greatly reliable.)



**Figure 1.** Electron dephasing rate  $\tau_\phi^{-1}$  as a function of temperature for three thick  $\text{Ag}_{40}\text{Pd}_{60}$  films having  $\rho(10\text{ K}) = 119$  ( $\square$ ), 178 ( $\circ$ ), and 281 ( $\triangle$ )  $\mu\Omega\text{ cm}$ . The solid curves are least-squares fits to equation (1).

Before the recent renewed interest in  $\tau_0$ , a saturation of the electron dephasing time as that shown in figure 1 has been observed in a good number of experiments, including reduced-dimensional metals [13] and semiconductors [14]. One of the most adopted (early) explanations proposed for the observed saturation invokes hot-electron effects. However, this kind of explanation can be ruled out for our measurements in figure 1. In the hot-electron picture, it is argued that if the Joule heating caused by measurement currents were sufficiently large, the conduction electrons would be removed out of thermal equilibrium with the phonon bath. Then, the electron temperature would be higher than the phonon (lattice) temperature, resulting in a ‘roll-over’ of  $\tau_\phi$  at the lowest measurement temperatures. Since we are concerned with three-dimensional systems in this work, Joule heating is *negligible* in our case. To confirm this assertion experimentally, we plot in figure 2 the variations of resistivity with temperature for those representative thick  $\text{Ag}_{40}\text{Pd}_{60}$  films just considered in figure 1. Comparison examination of figures 1 and 2 indicates that the resistivities of the samples keep

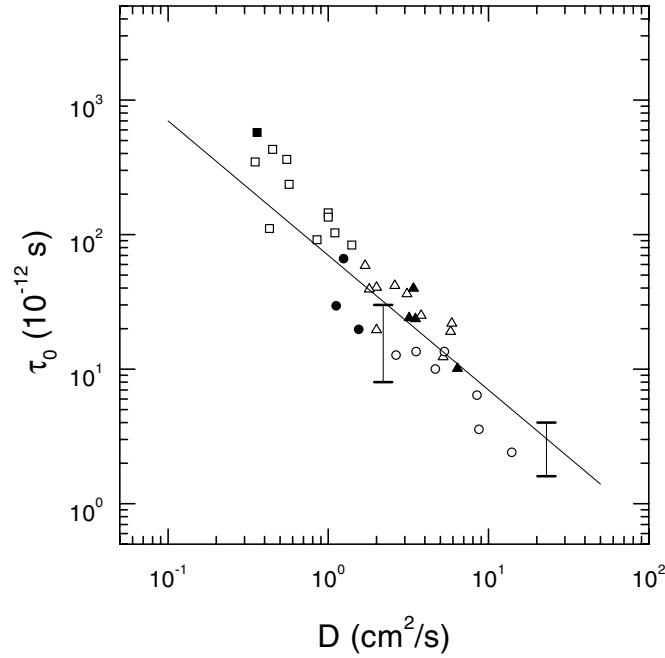
increasing with reducing temperature all the way down to our lowest measurement temperatures (400 mK), though a saturation in  $\tau_\phi$  is already seen at a much higher temperature of  $\sim 3$  K in these particular samples. Indeed, the resistivity rises with decreasing temperature shown in figure 2 can essentially be described by disorder enhanced electron–electron interaction effects [15], suggesting that the temperature of the electron gas is the same to the temperature of the lattice. Therefore, *no* appreciable Joule heating was produced in our measurements. Consequently, the saturation of  $\tau_\phi$  shown in figure 1 *cannot* be interpreted in terms of hot-electron effects. This is true for all samples studied.



**Figure 2.** Normalized resistivity  $\Delta\rho(T)/\rho(10\text{ K}) = [\rho(T) - \rho(10\text{ K})]/\rho(10\text{ K})$  as a function of  $\sqrt{T}$  for three thick  $\text{Ag}_{40}\text{Pd}_{60}$  films having  $\rho(10\text{ K}) = 119$  ( $\square$ ), 178 ( $\circ$ ), and 281 ( $\times$ )  $\mu\Omega\text{ cm}$ .

The central result of this work is plotted in figure 3. Figure 3 shows a plot of our measured saturated dephasing time  $\tau_0$  as a function of the electron diffusion constant  $D$  for more than 40 samples made from very different materials and prepared with very different fabrication techniques. Different symbols label different material systems as indicated in the caption to figure 3. In all cases, the samples are three-dimensional with regard to the electron phase coherence length  $\sqrt{D\tau_\phi}$ . Inspection of figure 3 reveals that the saturated dephasing time  $\tau_0$  for all samples falls essentially on the same region in the coordinates. Particularly, it reveals that all that matters in determining the value of  $\tau_0$  is the diffusion constant  $D$ , regardless of the distinct material characteristics (e.g. electronic structures) of the various samples. Close inspection of figure 3 indicates that  $\tau_0$  varies essentially with the diffusion constant  $D$  as  $\tau_0 \sim D^{-\alpha}$ , with  $\alpha$  close to or slightly larger than 1, for about *two decades* of  $D$ . As  $D$  increases from about 0.1 to 10  $\text{cm}^2\text{ s}^{-1}$  (corresponding to the quantity  $k_F l$  ranging from of order unity to of order 10, where  $k_F$  is the Fermi wave number, and  $l$  is the electron elastic mean free path),  $\tau_0$  decreases correspondingly from about 0.5 to 0.005 ns. In this figure, the straight solid line is drawn proportional to  $D^{-1}$  and is a guide to the eye. This observed  $\tau_0 \sim D^{-\alpha}$ , with  $\alpha$  close to or slightly larger than 1, is totally unexpected. On the contrary, it is often conjectured that  $\tau_0$  should increase with *reducing* disorder, *at least* in one and two dimensions [5]. Until now, it is *not* known exactly how differently the saturated dephasing time should behave in different dimensionalities and in different sample structures. Surprisingly, we notice

that our observation of an essentially inverse linear dependence of  $\tau_0$  on  $D$  actually implies an essentially ‘constant’ saturated dephasing length of  $\sqrt{D\tau_0} \sim 1000 \text{ \AA}$  in all samples. For comparison, the experimental values of  $\tau_0$  previously reported by Lin and Giordano [13] in thin, polycrystalline AuPd films are also indicated in figure 3 by the two vertical bars: the vertical bar at  $D \approx 2.2 \text{ cm}^2 \text{ s}^{-1}$  stands for the range of  $\tau_0$  observed in their dc sputtered films, while the vertical bar at  $D \approx 23 \text{ cm}^2 \text{ s}^{-1}$  stands for that in their thermally evaporated films. The observation of figure 3 is the first time such a systematic study has been done on so many samples with a wide range of diffusion constants.



**Figure 3.** Variation of saturated dephasing time  $\tau_0$  with electron diffusion constant  $D$  for various three-dimensional polycrystalline impure metals:  $\text{Au}_{50}\text{Pd}_{50}$  (circles),  $\text{Ag}_{40}\text{Pd}_{60}$  (squares),  $\text{Sb}$  (triangles),  $\text{Au}_x\text{Al}$  (solid triangles),  $\text{Sc}_{85}\text{Ag}_{15}$  (solid squares), and  $\text{V}_{100-x}\text{Al}_x$  (solid circles). The two vertical bars at  $D \approx 2.2$  and  $23 \text{ cm}^2 \text{ s}^{-1}$  represent the experimental values of  $\tau_0$  for dc sputtered and thermally evaporated thin, polycrystalline AuPd films, respectively, taken from [13]. The straight solid line is drawn proportional to  $D^{-1}$  and is a guide to the eye.

We discuss briefly how the values of the electron diffusion constant  $D$  for our samples are determined. For most of our samples, the values of  $D$  are computed through the Einstein relation  $D = [\rho_0 e^2 N(0)]^{-1}$ , using the measured values for the impurity resistivity  $\rho_0$ ; the values of the electronic density of states at the Fermi level  $N(0)$  are evaluated through measured low-temperature specific heats. While it is understood that the evaluation of  $D$  for some metal alloys could probably be subject to an uncertainty of as large as a factor  $\sim 2$ , due to the complex electronic structures, we stress that our experimental availability of a very wide range of  $D$  ensures that even with a factor of 2 in the uncertainty of  $D$  the main conclusion of the present work would not change.

One of the most popular (early) explanations for the saturation of  $\tau_\phi$  invokes spin–spin scattering due to the presence of a very minor amount of magnetic impurities in the sample. This explanation was refuted in many experiments [5, 13]. In the present work, this explanation

can also be ruled out simply because our samples were made from very different high-purity sources obtained from various reputable suppliers, and measured at very different times. It is hard to conceive that magnetic spin-spin scattering due to ‘accidental’ contamination could have caused the ‘systematic’ variation of  $\tau_0$  with  $D$  as observed in figure 3. Moreover, since we are concerned with bulk samples, any spin-flip scattering that might result from surface effects, such as interfaces, substrates, and paramagnetic surface oxidation, can largely be minimized (while surface effects could be significantly more important in reduced-dimensional systems). Indeed, as mentioned, we do *not* see a logarithmic Kondo type of dependence in resistivity (figure 2) in our temperature range, strongly ruling out magnetic impurities playing any role in our samples. It is also worthwhile to note that, although it has been widely taken for granted over several years that magnetic impurities will cause dephasing and produce a temperature independent  $\tau_\phi$ , Webb and coworkers [5] have recently ruled out magnetic impurities as the cause of the observed saturation.

The origin for the observed saturated dephasing time given in figure 3 is not exactly clear. To our knowledge, Altshuler *et al* [16] (see, also, [2]) have considered the dephasing of electron wave amplitudes by high-frequency electromagnetic noises. They found that an electromagnetic noise can be, on the one hand, already large enough to cause dephasing while, on the other hand, still too small to cause significant Joule heating of the conduction electrons. Unfortunately, in the most effective frequency range in causing dephasing, their theory predicts a form  $\tau_0 \sim D^{-1/3}$  which is in disagreement with our experimental observation. Besides, recent experiments [8,17] have explicitly demonstrated that direct dephasing due to radiation could not be the cause of the observed saturation. Therefore, we have to turn to other explanations, such as, among others, zero-point fluctuations of the electrons [1], dynamical two level systems [3], and coherent charge transfer between crystallites [18]. Unfortunately, in terms of the zero-point fluctuations theory, we observe, in almost all samples at 500 mK, a  $\tau_0$  already exceeds 3 to 5 orders in magnitude above the upper bound set by the theory. (The saturation time in three dimensions is given by the zero-point fluctuations theory as  $\tau_{GZ}^{(3D)} = (22\pi^2\hbar/e^2)(D^2/\rho_0 v_F^3)$ , where  $\rho_0$  is the residual impurity, and  $v_F$  is the Fermi velocity (see, e.g. [1,2]). Even worse, the zero-point fluctuations theory predicts increasing  $\tau_0$  with increasing  $D$ , opposite to our observation in *polycrystalline* impure metals. Although an increase of  $\tau_0$  with increasing mobility has been observed in, e.g. *semiconductor quantum wires* [19] and *quantum dots* [7,8], it is conceived [18] that the saturation behaviour in polycrystalline impure metals could be different from that in well-textured semiconductor quantum wires and dots. Indeed, it is our opinion that the saturation behaviour should be universal among polycrystalline impure metals, while different functional forms of  $\tau_0$  on disorder might be expected for different sample structures. If this were the case, then the current concept for zero-temperature dephasing would need major redirection.

Very recently, we have learned that, using *polycrystalline* thin AuPd wires and films, Natelson *et al* [20] have demonstrated that *sample geometry* alone could be essential for the saturation of electron dephasing time at  $T \rightarrow 0$ . They reported that, while saturation is absent in their one-dimensional wires down to their lowest measurement temperatures (80 mK), a relative saturation is clearly present as  $T \rightarrow 0$  for their two-dimensional films made of the same material. (The same experimental method and the same dilution refrigerator were used in their measurements for wires and films.) Their result is interesting. Taken together with the present observation of a pronounced  $\sqrt{D\tau_0} \sim 1000 \text{ \AA}$  in three-dimensional *polycrystalline* metals, our result and their measurements seem to point coherently to an experimental fact of *an enhancement of saturation of electron dephasing with increasing (higher) dimensions in polycrystalline metals*.

In conclusion, we have observed a saturated dephasing time  $\tau_0$  on the order of 0.005 to 0.5 ns in more than 40 three-dimensional polycrystalline impure metals with distinct characteristics. Taking many metals and alloys together, our result indicates that  $\tau_0$  varies essentially with the electron diffusion constant  $D$  as  $\tau_0 \sim D^{-\alpha}$ , with  $\alpha$  close to or slightly larger than 1, for about two decades of  $D$ . It is the first time such a systematic study has been done on so many samples with a wide range of diffusion constants. A complete theoretical explanation for this observation is not yet available. On the other hand, our result suggests that the origin for the observed saturation at low temperatures might not be universal over all dimensionalities and all kinds of sample structures. If this is the case, then the current concept for zero-temperature dephasing might need major redirection.

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