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Effect of annealing on electron dephasing in three-dimensional polycrystalline metals

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Abstract. – We have studied the effect of thermal annealing on electron dephasing times τ_ϕ in three-dimensional polycrystalline metals. Measurements are performed on as-sputtered and annealed AuPd and Sb thick films, using the weak-localization method. In all samples, we find that τ_ϕ possesses an extremely weak temperature dependence as $T \to 0$. Our results show that the effect of annealing is non-universal, and it depends strongly on the amount of disorder quenched in the microstructures during deposition. The observed "saturation" behavior of τ_ϕ cannot be easily explained by magnetic scattering. We suggest that the issue of saturation can be better addressed in three-dimensional, rather than lower-dimensional, structures.

Introduction. – The electron dephasing time τ_ϕ is a very important quantity that governs the mesoscopic phenomena at low temperatures. Particularly, the behavior of the dephasing time near zero temperature, $\tau_\phi^0 = \tau_\phi(T \to 0)$, has recently attracted vigorous experimental [1–7] and theoretical [8–13] attention. One of the central themes of this renewed interest is concerned with whether τ_ϕ^0 should reach a finite or an infinite value as $T \to 0$. The connection of the zero-temperature dephasing behavior with the very fundamental condensed-matter physics problems such as the validity of the Fermi-liquid picture, the possibility of the occurrence of a quantum phase transition, and the persistent currents in metals, etc., has been addressed. Conventionally, it is accepted that τ_ϕ^0 should reach an infinite value if there exist only the inelastic electron-electron and electron-phonon scattering. However, several recent measurements performed on different mesoscopic conductors have revealed that τ_ϕ^0 depends only very weakly on T, if at all, when T is sufficiently low. There is no generally accepted process of electron–low-energy-excitation interactions that can satisfactorily explain the "saturation" of τ_ϕ^0 found in the experiments. It should be noted that those experiments

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Table I – *Resistivities, diffusion constants, and dephasing times of the as-sputtered* AuPd *and* Sb *thick films. The value of* τ_ϕ^0 *is extracted by least-squares fitting the measured* $\tau_\phi(T)$ *to eq.* (1).

Film	ρ(300 K) ($\mu\Omega$ cm)	ρ_0 ($\mu\Omega$ cm)	D (cm^2/s)	τ_ϕ^0 (10^{-10} s)
AuPd1e	131	124	4.9	0.18
AuPd4a	509	467	1.3	0.85
AuPd5e	535	473	1.3	0.88
AuPd6e	117	115	5.3	0.14
Sb01B	701	746	5.8	0.24
Sb12	1485	1645	2.6	0.51

[1,3,4,6,7] have ruled out electron heating, external microwave noises, and very dilute magnetic impurities as the origins for the observed finite dephasing as $T \to 0$.

To unravel the issue of electron dephasing, systematic information of τ_ϕ^0 over a wide range of sample properties is very desirable. Bearing this in mind, we have in this work performed systematic measurements of τ_ϕ^0 on several series of *as-sputtered* and subsequently *annealed* AuPd and Sb *thick* films. The low-field magnetoresistances of the as-sputtered samples are first measured. The samples are then annealed, and their magnetoresistances measured. The annealing and magnetoresistance measurement procedures are repeated a few times. τ_ϕ is extracted by comparing the measured magnetoresistances with the three-dimensional (3D) weak-localization (WL) theoretical predictions [14]. Generally, thermal annealing causes a decrease in the sample resistivity, signifying a reduction in the amount of defects in the microstructures. Controlled annealing measurements are thus crucial for testing the theoretical models of electron dephasing invoking magnetic impurities and dynamical defects [15].

Experimental method. – Thick-film samples were prepared by dc sputtering deposition onto glass substrates held at room temperature. The deposition rate was varied to tune the amount of disorder, *i.e.*, the residual resistivity ρ_0 (= ρ(10 K)) of the films. The AuPd films were typically 6000 Å × 0.3 mm × 17 mm, while the Sb films were typically 3000 Å ×0.3 mm × 17 mm. Thermal annealing of the AuPd (Sb) films was performed in a 99.999% pure Ar atmosphere at moderate temperatures of ∼ 100–300 °C (∼ 150 °C) for about one-half to several hours until ρ_0 changed by a desirable amount. The use of an extremely high-purity Ar atmosphere greatly minimized the presence of any oxygen residual gas in the annealing. The values of the relevant parameters for our as-sputtered films are listed in table I.

We notice that the four AuPd films listed in table I were *newly* made from a *new* $Au_{50}Pd_{50}$ target different from that used in our previous study [3]. Moreover, a *different* sputtering gun and a different vacuum chamber were utilized. Previously, we had studied τ_ϕ in a series of dc sputtered AuPd thick films prepared and measured in a different laboratory [3, 16]. By applying these new samples, we are able to perform a close comparison study of τ_ϕ in the same material prepared under different conditions. Such a comparison is indispensable for clarifying the possible role, if at all, of magnetic scattering on τ_ϕ^0. If there were any noticeable magnetic contamination during this experiment, it is natural to expect an *unintentional* magnetic concentration, $n_{\rm m}$, that differs from that in our previously samples [3]. Consequently, a distinct value of τ_ϕ^0 should be observed. On the other hand, if a similar value of τ_ϕ^0 is measured, this result must then bear important information about an intrinsic material property.

In addition to the newly prepared AuPd samples, we have studied two "aged" Sb films. The two Sb films listed in table I were first deposited and studied two years ago in ref. [17]. During the past two years, they were exposed to air all the time. One might have naively speculated that these two samples must be heavily contaminated by (magnetic) impurities and, thus,

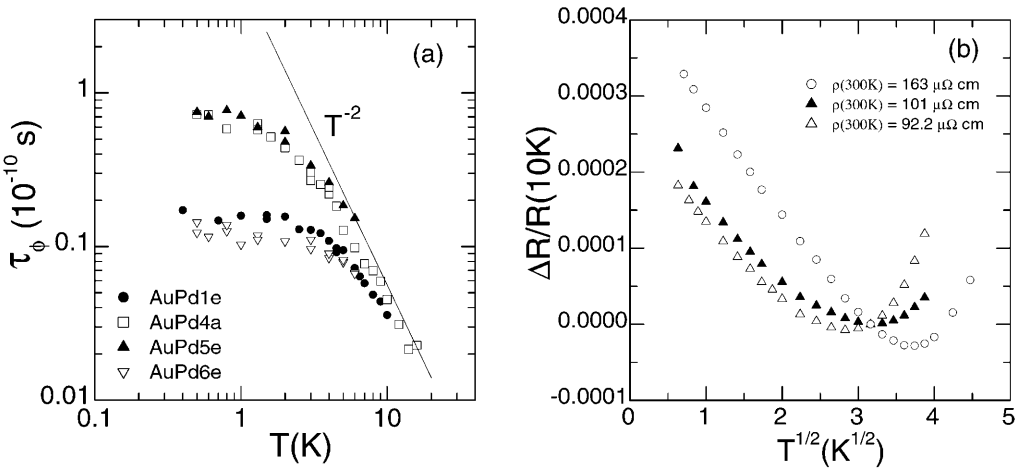

Fig. 1 – (a) τ_ϕ as a function of temperature for four as-prepared AuPd thick films. (b) $\Delta R/R(T) = [R(T) - R(10\,\mathrm{K})]/R(10\,\mathrm{K})$ as a function of $\sqrt{T}$ for three AuPd thick films.

have a shorter τ_ϕ with a much weaker T-dependence, compared with that measured two years ago. To the contrary, this experiment indicates that 3D AuPd and Sb are *not* as vulnerable to contamination as speculated. Our results point to an experimental fact that suggests that the observed saturation of τ_ϕ^0 cannot be readily explained by magnetic scattering.

Results and discussion. – We have measured the magnetoresistances and compared with 3D WL predictions [14] to extract the values of τ_ϕ. Our experimental method and data analysis procedure had been discussed previously [18]. Here we emphasize that, in the limit of strong spin-orbit scattering (which applies for both AuPd and Sb), τ_ϕ is the *only* adjusting parameter in the least-squares fits of the measured magnetoresistances with WL predictions. This great advantage makes the extraction of τ_ϕ highly reliable. Empirically, τ_ϕ can be written in the form

$$1/\tau_\phi(T) = 1/\tau_\phi^0 + 1/\tau_\mathrm{i}(T)\,, \tag{1}$$

where τ_ϕ^0 dominates at the lowest measurement temperatures, and τ_i is the relevant inelastic scattering time which is usually important at a (few) degree(s) kelvin and higher. In three dimensions, electron-phonon scattering is the predominant inelastic process while the Nyquist electron-electron scattering is negligibly small [19], *i.e.* $1/\tau_\mathrm{i} \approx 1/\tau_\mathrm{ep}$ in eq. (1). The electron-phonon scattering rate $1/\tau_\mathrm{ep}$ varies as T^p, with $2 \leq p \leq 4$.

Figure 1(a) shows our measured τ_ϕ as a function of temperature for four as-sputtered AuPd films. This figure demonstrates that τ_ϕ first increases with decreasing T at a few degrees kelvin, where the electron-phonon scattering dominates the total dephasing and $1/\tau_\phi \approx 1/\tau_\mathrm{ep} \propto T^2$ in AuPd [16]. Below about 2 K, the inelastic process is much less effective and a new mechanism progressively takes over, resulting in a very weak temperature dependence of τ_ϕ as $T \to 0$. To our knowledge, there is no generally accepted process of electron–low-energy-excitation interactions that can account for such a weak T behavior. A weak temperature dependence of τ_ϕ^0 is also observed in the two Sb thick films listed in table I. In fact, an (almost) absence of temperature dependence of τ_ϕ has previously been found in numerous three-dimensional polycrystalline metals [3]. We notice that the values of τ_ϕ^0 in fig. 1(a) follow *closely* the scaling relation of $\tau_\phi^0 \approx 10^{-10} D^{-1}$ s established in fig. 3 of [3], where the diffusion constant D is in

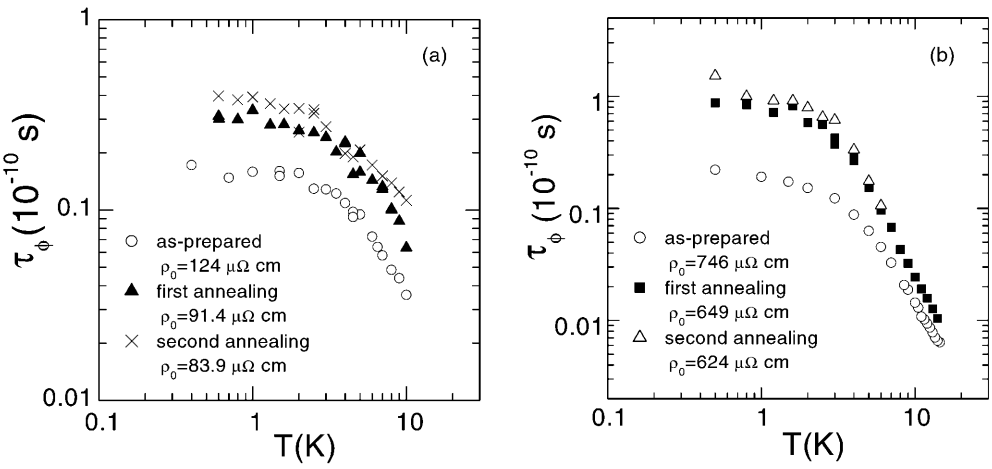

Fig. 2 – τ_ϕ as a function of temperature for (a) the as-prepared and annealed AuPd1e thick film, and (b) the as-prepared and annealed Sb01B thick film.

cm^2/s. This result suggests that the behavior of τ_ϕ found in fig. 1(a) is material intrinsic.

It should be emphasized that the weak T-dependence or the so-called "saturation" of τ_ϕ^0 in fig. 1(a) is observed in a temperature regime where the sample resistance varies as $-\sqrt{T}$ all the way down to our lowest measurement temperatures (fig. 1(b)). This $(-\sqrt{T})$-dependence of the resistance is well described by the 3D electron-electron interaction effects [20]. This result indicates that the saturation of τ_ϕ^0 is *not* caused by electron heating. A similar assertion of non–hot-electron effects has also been reached in previous experiments [1, 3, 4, 6, 7].

Information about the effect of annealing is crucial for clarifying the nature of magnetic scattering and dynamical defects. Figure 2(a) shows the variation of τ_ϕ with temperature for the as-prepared and subsequently annealed AuPd1e thick film. This figure clearly indicates that τ_ϕ increases with annealing. Similar behavior of increasing τ_ϕ with annealing has also been found in the as-prepared and annealed AuPd6e thick film. At first glance, this observation is easily explained. Suppose that annealing results in the rearrangement of lattice atoms and relaxation of grain boundaries and, hence, makes the film less disordered. Because two-level systems (TLS) are closely associated with the presence of defects in the microstructures, their number concentration would be reduced by annealing. By assuming that dynamical defects are effective scatterers, one can then understand fig. 2(a) in terms of a reducing TLS picture. However, our further measurements indicate that the nature of low-temperature dephasing in real metals is not so straightforward. We find that the effect of annealing on τ_ϕ is distinctly different in strongly disordered samples (fig. 3). For the convenience of discussion, the AuPd1e and AuPd6e (AuPd4a and AuPd5e) thick films are referred to as moderately (strongly) disordered, because they have $\rho_0 \sim 120$ (470) $\mu\Omega\,$cm before annealing.

Figure 2(b) shows the variation of τ_ϕ with T for the as-prepared and annealed Sb01B thick film. This figure clearly indicates that τ_ϕ increases with annealing. Similar effect of annealing has also been found in the Sb12 thick film. The results of figs. 2(a) and (b) suggest that an enhancement of τ_ϕ by thermal annealing is common to different *moderately* disordered metals. (We notice that the high resistivities in Sb films arise from a low carrier concentration instead of a short electron mean free path [17]. Our Sb thick films are thus moderately disordered.)

The importance of three-dimensional structures. – One of the widely accepted explanations for the "saturation" behavior of τ_ϕ^0 invokes magnetic spin-spin scattering due to a low-level contamination of the sample. This explanation has been challenged in several recent careful experiments [1, 3, 4, 6, 7]. However, despite this experimental situation, there is still an insisting opinion that argues for non-zero magnetic scattering in the sample. To completely reject such an opinion is non-trivial, because it is argued that the level of unintentional contamination is so low that it cannot be detected by the state-of-the-art material analysis techniques. The situation becomes more serious when reduced-dimensional systems are involved. In the case of low-dimensional structures, surface effects due to interfaces, substrates, and paramagnetic oxidation are non-negligible. Therefore, it is not straightforward to ascribe the observed saturation of τ_ϕ^0 to either intrinsic material properties or surface/interface effects. On the other hand, this kind of ambiguity does *not* occur in our *three-dimensional* measurements. In fact, we believe that magnetic scattering can at most play a subdominant role in our experiment. Our reasons are given as follows. i) Suppose that there is a low level of magnetic contamination in our as-sputtered films. Upon annealing, the magnetic impurity concentration $n_{\rm m}$ should be left unchanged. If the original "saturation" in our as-sputtered samples is caused by spin-spin scattering, one should then expect the same value of τ_ϕ^0 ($\propto n_{\rm m}^{-1}$) after annealing. However, we find increasing τ_ϕ^0 with annealing. Our result is thus in disagreement with this assumption. ii) Our argument for a non-magnetic origin is supported by the observation of an *increased* τ_ϕ^0 in the aged and annealed Sb films. Since our Sb01B and Sb12 thick films were aged in air for two years, one might have naively expected a large decrease in τ_ϕ^0 due to magnetic contamination. Nevertheless, this is not the case found in fig. 2(b). iii) Moreover, if our samples do contain an appreciable level of unintentional magnetic impurities, the contaminated concentration $n_{\rm m}$ should be basically the same in all films, because similar fabrication and measurement procedures were involved. One should then expect a similar τ_ϕ^0 in all *as-prepared* samples, regardless of disorder. This is certainly inconsistent with the observed scaling relation $\tau_\phi^0 \propto D^{-1}$ discussed above. Therefore, magnetic scattering in its current form cannot easily explain our overall results in a consistent manner.

In order to explain the widely observed saturation behavior of τ_ϕ^0, it has recently been proposed that dynamical defects can be important [10, 11]. The low-energy excitations of the dynamical defects are usually modelled by two-level systems. We already discussed that TLS might be partly responsible for the saturated dephasing found in our moderately disordered films. However, it is impossible to perform a quantitative comparison of our experiment with the TLS theories. The difficulties lie on the facts that i) the number concentration of TLS in a particular sample is not known, ii) the strength of coupling between conduction electrons and a TLS is poorly understood, and iii) the dynamical properties of real defects (impurities, grain boundaries, etc.) are even less clear. Experimentally, we also find other features of thermal annealing (fig. 3) that seem incompatible with a TLS picture of dephasing.

In addition to the moderately disordered samples, we have performed measurements on thick films containing much higher levels of disorder. Surprisingly, we discover that the effect of annealing is completely different. In the *strongly* disordered AuPd4a and AuPd5e thick films, we find that annealing causes a *negligible* effect on τ_ϕ. Figure 3 shows the variation of τ_ϕ with T for the as-prepared and annealed AuPd4a thick film. This figure clearly demonstrates that the values of τ_ϕ for the as-prepared and annealed samples are essentially the same, even though the resistance, and hence diffusion constant D changed by a factor of more than 6. The absence of an appreciable annealing effect in this case implies that, in addition to the usual TLS addressed above, these two films contain other defects that cannot be readily cured by thermal annealing. Such a null effect of annealing seems to suggest that, despite a large effort

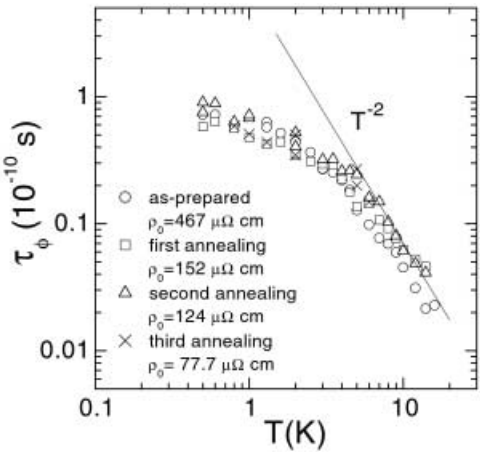

Fig. 3 – τ_ϕ as a function of temperature for the as-prepared and annealed AuPd4a thick film.

in this direction, no real defects of any nature can be found to have dynamical properties that may explain the saturation behavior of τ_ϕ^0. A comparison of figs. 2 and 3 strongly indicates that low-temperature dephasing is very sensitive to the microstructures.

The observation of fig. 3 deserves further discussion. First, we recall that our measured τ_ϕ in the as-sputtered, strongly (and moderately) disordered films follows the scaling relation of $\tau_\phi^0 \propto D^{-1}$ mentioned above [3], implying that the result of fig. 3 is material intrinsic. Secondly, in the context of magnetic scattering, Blachly and Giordano recently found that Kondo effect is very *sensitive* to disorder, namely, an increase in disorder suppresses Kondo effect [21]. Along this line, if the original saturated τ_ϕ^0 found in fig. 3 were due to magnetic scattering, one should argue that thermal annealing that suppresses disorder should enhance Kondo effect. Then, a decreased τ_ϕ^0 should be expected with annealing. Since our τ_ϕ^0 does not change even when the sample resistivity is reduced by a factor of more than 6 by annealing, fig. 3 thus cannot be easily reconciled with a magnetic scattering scenario. Besides, this picture of a weakened Kondo effect by disorder is also incompatible with our result for the moderately disordered films (fig. 2) where an increased, instead of a decreased, τ_ϕ^0 with annealing is found. Thirdly, one might argue that annealing in the presence of oxygen can lead to the oxidation of magnetic impurities, and hence to the disappearance of Kondo resistivity [22]. We argue that this is unlikely in our case, since we had taken precautions by annealing our films in a 99.999% pure Ar atmosphere, where the amount of oxygen residual gas was greatly minimized. Moreover, since we do not find a rapid increase in the T-dependence of τ_ϕ^0 after annealing, there is thus no clear evidence of the presence of oxygen or magnetic impurities in our films. In any case, it would be interesting to study if annealing in oxygen would have a drastic effect on τ_ϕ^0.

Lastly, we discuss the advantage of using three-, instead of lower-dimensional, mesoscopic structures for τ_ϕ measurements. In 3D, the dominating inelastic process is the electron-phonon scattering for which $\tau_{\rm ep}$ obeys a strong T^{-p} ($2 \leq p \leq 4$) dependence [16, 19]. Such a temperature variation is much stronger than the dominating $p = 2/3$ in one dimension and $p = 1$ in two dimensions (which are both due to electron-electron scattering [20]). Inspection of the solid lines, which are drawn proportional to T^{-2}, in figs. 1 and 3 reveals that our measured τ_ϕ^0 at 0.3 K is already $\sim$ *two orders of magnitude* lower than that as would be extrapolated from the measured $\tau_{\rm ep}$ at a few degrees kelvin. Such a huge discrepancy is well outside any

experimental uncertainties. On the contrary, in the case of narrow wires, the dominating inelastic dephasing time obeys a much weaker $T^{-2/3}$ law just mentioned. In this case, any discrepancy between the measured and extrapolated values of τ_ϕ^0 would be less dramatic in the attainable experimental temperature range, rendering a discrimination of the presence or absence of a saturated τ_ϕ^0 less clear-cut.

Conclusion. – We have studied the influence of thermal annealing on low-temperature electron dephasing in polycrystalline AuPd and Sb thick films. We find that τ_ϕ^0 reveals an extremely weak temperature dependence in both as-sputtered and annealed samples. The effect of annealing is non-universal, depending strongly on the amount of disorder quenched in the microstructures during deposition. The observed saturation behavior of τ_ϕ^0 cannot be easily explained by magnetic-scattering in its current form. We also find that the disorder behavior of τ_ϕ^0 in as-prepared and annealed samples is very different. A complete theoretical explanation would need to take the microstructures into account.

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