

Observation of Strong Electron Dephasing in Highly Disordered $\text{Cu}_{93}\text{Ge}_4\text{Au}_3$ Thin Films

S. M. Huang,^{1,2} T. C. Lee,³ H. Akimoto,⁴ K. Kono,² and J. J. Lin^{1,3,*}

¹*Institute of Physics, National Chiao Tung University, Hsinchu 30010, Taiwan*

²*Low Temperature Physics Laboratory, RIKEN, Hirosawa 2-1, Wako-shi, Saitama 351-0198, Japan*

³*Department of Electrophysics, National Chiao Tung University, Hsinchu 30010, Taiwan*

⁴*Nano-Science Joint Laboratory, RIKEN, Hirosawa 2-1, Wako-shi, Saitama 351-0198, Japan*

(Received 15 November 2006; published 26 July 2007)

We report the observation of strong electron dephasing in a series of disordered $\text{Cu}_{93}\text{Ge}_4\text{Au}_3$ thin films. A very short electron dephasing time possessing very weak temperature dependence around 6 K, followed by an upturn with further decrease in temperature below 4 K, is found. The upturn is progressively more pronounced in more disordered samples. Moreover, a $\ln T$ -dependent, but high-magnetic-field-insensitive, resistance rise persisting from above 10 K down to 30 mK is observed in the films. These results suggest a nonmagnetic dephasing process which is stronger than any known mechanism and may originate from the coupling of conduction electrons to dynamic defects.

DOI: 10.1103/PhysRevLett.99.046601

PACS numbers: 72.10.Fk, 73.20.Fz, 73.23.-b

The electron dephasing time τ_φ is the key quantity in the mesoscopic physics. Recently, intense theoretical [1–5] and experimental [6–9] efforts have been made to address the low temperature behavior of τ_φ . In particular, the issue of whether the value of τ_φ should saturate or diverge as $T \rightarrow 0$ K is being discussed. A “saturated” τ_φ would imply a breakdown of the Fermi-liquid picture in a mesoscopic system at very low T [6]. The inelastic scattering between the conduction electrons and a localized moment in a metal host containing dilute magnetic impurities has recently been intensely studied in this context [9–14]. It has also been argued if intrinsic electron processes might lead to a saturation in $\tau_\varphi(T \rightarrow 0)$ [1]. In addition, there are theories suggesting that materials properties associated with dynamical structural defects may cause profound dephasing at low T [2–5]. Despite these different proposals, it is accepted that the responsible electron dephasing processes in (comparatively) highly and weakly disordered metals are dissimilar [5,8]. That is, while it is realized that magnetic scattering is important in weakly disordered metals [10], a different mechanism is believed to be relevant for the weak T dependence of τ_φ found in more disordered alloys [15–19]. Thus, to understand the issue of the interactions between conduction electrons and low-lying excitations, and, more importantly, to resolve the low- T dephasing problem, information about τ_φ in highly disordered metals is indispensable. In this work, we have measured τ_φ as a function of T and the sheet resistance R_\square in a series of two-dimensional (2D) $\text{Cu}_{93}\text{Ge}_4\text{Au}_3$ thin films. Our values of the electron diffusion constant $D (\propto \rho^{-1})$, ρ is the resistivity) are a factor approximately several tens smaller than those ($D \approx 100\text{--}200 \text{ cm}^2/\text{s}$) in the metal wires recently studied in Refs. [6,8,13,14]. Our results provide strong evidence that the origin for the saturation in τ_φ is nonmagnetic.

We have chosen $\text{Cu}_{93}\text{Ge}_4\text{Au}_3$ (CuGeAu for short) as our system material. Ge atoms were doped into the Cu host

mainly to increase the impurity scattering to enhance the weak-localization (WL) effects, while Au atoms were doped mainly to introduce spin-orbit scattering. In the limit of strong spin-orbit scattering, τ_φ becomes the only free parameter in the comparison of the experimental magnetoresistances (MRs) with the WL theory [20], making the extraction of τ_φ highly reliable. (Good fits of the 2D WL theories to our measured perpendicular MRs were obtained for all films, and are shown for a representative film in Fig. 1.) The starting CuGeAu target was intentionally chosen to be only of a “medium” purity (99.99%) while, on the other hand, the spectrographic analysis of the target indicated low levels of 4 (Fe), 0.3 (Mn), and 0.003 (Cr) ppm of magnetic impurities.

Our films were made by dc sputtering deposition on glass substrates held at ambient temperature. The deposition rate was adjusted to tune the level of disorder of the

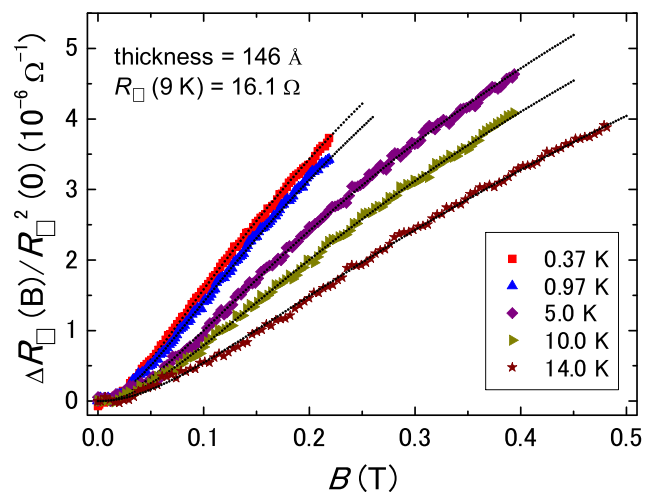


FIG. 1 (color online). Perpendicular MRs for a CuGeAu thin film at several temperatures as indicated. The dotted curves are the 2D WL theoretical predictions.

films. The films were of meander shape, defined by either a mechanical mask or photolithography, with typical length of ≥ 5.7 mm, width of ~ 0.1 mm, and thickness of either $\approx 147 \pm 3$ or 195 ± 5 Å. Our values of $R_{\square}(9\text{ K})$ varied from 9.14 to 62.9 Ω , corresponding to $D \approx 4\text{--}25$ cm²/s and $k_F l \approx 10\text{--}65$ (k_F is the Fermi wave number, and l is the electron mean free path). Here $D = v_F l / 3$ was evaluated using the Fermi velocity $v_F(\text{Cu}) = 1.57 \times 10^6$ m/s, and l was computed using $\rho l = 6.4 \times 10^{-6}$ $\mu\Omega$ cm² for Cu. The resistance ratio $R_{\square}(300\text{ K})/R_{\square}(9\text{ K}) = 1.1042\text{--}1.1109$. For complementariness, we have also studied a few 3D (≥ 0.5 μm thick) films made from the same target, whose resistivity ratio $\rho(300\text{ K})/\rho(10\text{ K}) = 1.047\text{--}1.070$. Such low values of residual resistance ratios reflect that our sputtered samples must contain large amounts of (structural) defects. To avoid electron overheating, the condition for equilibrium $eV_{\mathcal{E}} \ll k_B T$ was assured in all resistance and MR measurements, where $V_{\mathcal{E}}$ is the applied voltage across the energy relaxation length [17].

Figure 2 shows the main result of this work, i.e., the measured τ_{ϕ} as a function of T for a series of thin films with different values of R_{\square} . (Notice that the values of τ_{ϕ} for each film have been vertically shifted for clarity.) One of the most distinct features revealed in Fig. 2 is that τ_{ϕ} possesses a very weak T dependence around 6 K, being

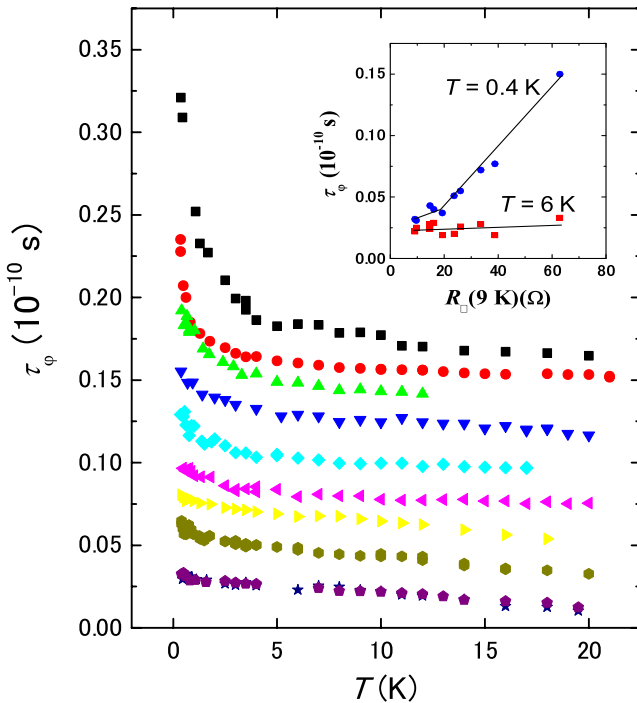


FIG. 2 (color online). τ_{ϕ} as a function of T for CuGeAu thin films with $R_{\square}(9\text{ K})$ (from bottom to top) = 9.14, 9.66, 14.7, 16.1, 19.3, 23.7, 26.0, 33.6, 38.8, and 62.9 Ω . Except for the lowest two curves, each curve has been consecutively shifted up by a value of 0.002 ns from its lower-neighbor curve. The inset shows the variation of the measured $\tau_{\phi}(0.4\text{ K})$ and $\tau_{\phi}(6\text{ K})$ with $R_{\square}(9\text{ K})$. The solid lines are guides to the eye.

very short, and has a very similar magnitude in this “plateau” regime. The inset indicates an almost constant $\tau_{\phi}(6\text{ K}) \approx 0.002\text{--}0.003$ ns for all films. The second distinct feature is that there is an upturn in τ_{ϕ} with decreasing T below about 4 K. In particular, the upturn is sample dependent, being progressively stronger in dirtier films. The inset shows our measured values of $\tau_{\phi}(0.4\text{ K})$ and $\tau_{\phi}(6\text{ K})$ versus $R_{\square}(9\text{ K})$. It depicts that $\tau_{\phi}(0.4\text{ K})$ monotonically increases with increasing disorder. Such upturn behavior is not seen in those recent measurements on weakly disordered samples [6,8,13,14]. Thirdly, the electron-phonon (e -ph) scattering rate $\tau_{e\text{-ph}}^{-1} \propto T^2$ [21] is found to be important only at $T \geq 10$ K. On the other hand, we note that the 2D electron-electron (e - e) scattering rate $\tau_{e\text{-}e}^{-1}$, which is often the dominant dephasing process in thin films at low T [22], is 2–3 orders of magnitude smaller than the measured τ_{ϕ}^{-1} shown in Fig. 2, and thus can be ignored in the following analysis [23].

As a first check of the T dependence of our measured τ_{ϕ} below 5 K, we write an effective power law $\tau_{\phi}^{-1} \propto T^p$ and compare it with our data to extract the value of p . We found that, even in the two most disordered films [$R_{\square}(9\text{ K}) = 38.8$ and 62.9 Ω] where the upturn is most profound, the value of p is still small ($\approx 0.57 \pm 0.06$). In other less disordered films, p is even much smaller, being close to zero in our cleanest films. In all cases, our values of p are markedly lower than that ($p = 1$) recently predicted in the theory for soft local defects induced dephasing at temperatures below the plateau regime [5].

Concerning the observation of a very weak T -dependent τ_{ϕ} , one immediately suspects if such behavior might be due to spin-spin scattering in the presence of dilute magnetic impurities. As a quick check, we notice that Cu could possibly contain trace Cr, Mn, or Fe impurities and form Kondo alloys. In a Kondo system, the magnetic scattering rate τ_m^{-1} is maximum at $T = T_K$, where also a plateau in the dephasing time $\tau_{\phi}^{\text{plateau}}$ often appears. Based on the Nagaoka-Suhl (NS) expression [24], Pierre *et al.* [8] pointed out that for Cu $\tau_{\phi}^{\text{plateau}} = (c_m / \pi \hbar \nu)^{-1} \approx (0.6 / c_m)$ ns, where c_m is the magnetic impurity concentration in parts per million (ppm), and ν is the total electron density of states at the Fermi level. Then, our experimental value of $\tau_{\phi}(6\text{ K}) \approx 0.002\text{--}0.003$ ns would imply a level of $c_m \approx 200\text{--}300$ ppm in the samples, if our measured τ_{ϕ}^{-1} were directly ascribed to τ_m^{-1} . Such a level of c_m is obviously too high to be realistic.

Recently, new theories have been established and it is realized that the NS result for τ_m^{-1} need be revised. The new calculations of Zarand *et al.* [10] and Micklitz *et al.* [11] for spin $S = \frac{1}{2}$ impurities have confirmed that the scattering rate τ_m^{-1} is maximum at $T = T_K$. Their corrected peak scattering rate is $\approx 8\%$ lower than the NS prediction. So the estimate of c_m given above will not be substantially altered even if one applies the new theory. In fact, for all T , the Zarand-Micklitz (ZM) theory predicts a τ_m^{-1} below the

corresponding NS scattering rate. The latter is in turn below our experimental scattering rate (see Fig. 3).

To unravel the intriguing dephasing mechanism responsible for our measured τ_φ , we carry out further quantitative analysis below. Assume that our measured τ_φ^{-1} between 0.3 and 25 K is given by $\tau_\varphi^{-1} = A_{e-ph}T^2 + \tau_Q^{-1}$ (recall that τ_{e-e}^{-1} is totally negligible), where τ_Q^{-1} denotes a yet-to-be identified dephasing rate, and the e -ph coupling strength A_{e-ph} can be determined from the high- T part of the measured τ_φ [21]. In Fig. 3, we plot the variation of τ_Q^{-1} with T for two representative films. Figure 3 clearly reveals a maximum in τ_Q^{-1} at a characteristic temperature which we denote as T_K^* . (We will see later that this T_K^* cannot be identified with the Kondo temperature T_K .) Below T_K^* , one sees that the T dependence of τ_Q^{-1} is much stronger for the higher R_\square film, as can be expected from the above discussion that the exponent p decreases with decreasing R_\square .

Since at $T \geq T_K$ the ZM theory [10,11] essentially reproduces the conventional wisdom, we first compare our experimental τ_Q^{-1} with the NS expression [24,25] for the temperature range $T \geq T_K^*$. In plotting the NS approximations in Fig. 3, we have adjusted the free parameters (T_K^* , c_m , and the local spin S) so that the theory reproduced the experiment at $T \geq T_K^*$. Figure 3 illustrates that the NS expression can describe our measurements down to slightly below T_K^* . However, inspection of the fitted values indicates that such agreement is spurious, because such good fits can only be achieved by using unrealistic values for the adjusting parameters. For example, a local spin of $S = 0.12$ (0.082) and $T_K^* = 7.2$ (4.75) K had to be used for the $R_\square(9\text{ K}) = 38.3$ (16.1) Ω film. Using $S = \frac{1}{2}$ or any larger value can never reproduce our data. Moreover, if we ascribe the measured $(\tau_Q^{-1})^{\max}$ to $\tau_m^{-1}(T = T_K)$, an unreasonably large value of $c_m = 200$ –300 ppm will be inferred, implying that an unusually strong τ_Q^{-1} is entirely dominating over the e -ph and e - e scattering in our films in this T range. Although accidentally formed CuO on the film surfaces may have $S = 1$ spin [26], one would not expect a huge $c_m > 200$ ppm to result from such oxidation. Besides, there are no known Cu-based Kondo alloys which have values of T_K around ≈ 5 –7 K. Thus, in all aspects, it is

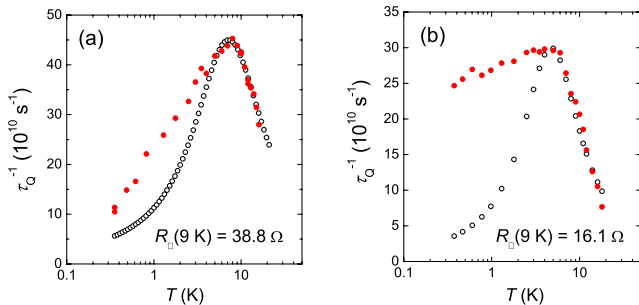


FIG. 3 (color online). τ_Q^{-1} as a function of T for two CuGeAu thin films. The solid symbols are the experimental data and the open symbols are the Nagaoka-Suhl expression; see text.

certainly impossible to ascribe our measured τ_Q^{-1} to magnetic scattering.

Another decisive way of checking whether magnetic impurities might exist to a degree in the samples is to explore if the variation of resistance with T reveals any Kondo or spin-glass behavior. Figure 4(a) shows the variation of R_\square with T for a representative thin film in zero field and in a high perpendicular magnetic field B . This figure clearly indicates that, both in $B = 0$ and 9 T, the R_\square reveals a $\ln T$ behavior all the way down to 50 mK. There is not even a sign of a crossover to a saturation characterizing a Kondo system in the unitary limit [13,27]. In fact, if there were $c_m \geq 200$ ppm in the films, we should have been in a spin-glass state at low $T < T_K$ and a marked decrease in R_\square should be observed as the local moments freeze into a collective state [28]. Moreover, in the presence of a large magnetic field $B \gg k_B T / g\mu_B$ (μ_B is the Bohr magneton), the local spins should be aligned and a notable decrease in R_\square should occur at low T [28]. However, none of these features are found in Fig. 4(a). Thus, our R_\square as a function of T and B does not support a picture based on magnetic impurities.

Usually, a $\ln T$ increase in R_\square in thin metal films may be due to WL and e - e interaction (EEI) effects, and the resistance rise can be written as $\Delta R_\square(T)/R_\square^2 = -\alpha_T(e^2/2\pi^2\hbar)\ln T$, where $\alpha_T = \alpha p + (1 - F')$. The parameter αp characterizes the WL effect and $1 - F'$ the EEI effect. For disordered metal films in the limit of strong spin-orbit interaction, the screening factor F' is small (typically, ≈ 0.1) and the EEI effect dominates the resist-

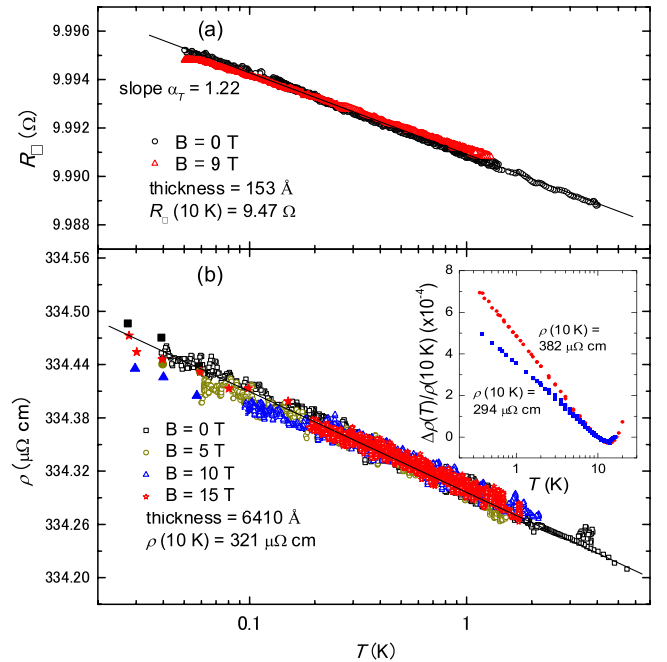


FIG. 4 (color online). (a) Variation of R_\square with T for a thin film. (b) Variation of ρ with T for a thick film. Inset: $\Delta\rho(T)/\rho(10\text{ K}) = [\rho(T) - \rho(10\text{ K})]/\rho(10\text{ K})$ as a function of T for two thick films.

ance rise while the WL contribution is negligibly small [22]. Then, these two effects would cause a maximum resistance rise with a slope $\alpha_T = 1$. For all the thin films studied in this work, we obtain $\alpha_T = 1.33 \pm 0.13$. This value is systematically larger than unity, strongly implying that there must be an extra mechanism which also contributes to the $\ln T$ rise in R_{\square} . This additional contribution is, however, insensitive to high magnetic fields.

To further illuminate this anomalous $\ln T$ resistance rise, we have also made a few thick films from the same sputtering target under similar deposition conditions and measured the resistance. Figure 4(b) shows the variation of ρ with T for a representative thick film in zero field and in perpendicular magnetic fields. Usually, for a disordered bulk metal, one expects to see a resistance rise obeying a $-\sqrt{T}$ law [22]. This figure, however, reveals a strict $\ln T$ behavior from above 10 K (see inset) down to 30 mK, indicating some mechanism being dominating over the 3D EEI effect in our samples. Moreover, there is definitely no sign of a saturated (decreased) resistance signifying the presence of the Kondo (spin-glass) behavior. Also, there is no evidence of profound negative MRs in the presence of a high B . These results strongly suggest that magnetic scattering, if any exists, could not be the primary mechanism in our samples. On the other hand, it has recently been reported that scattering of electrons off two-level systems can cause a $\ln T$ and high- B -insensitive variation of the resistance down to very low temperatures [29]. Our results in Fig. 4 are in line with this observation. If dynamic defects somehow abound in our sputtered films, then it is natural to ask to what extent such defects can contribute to dephasing [2,3,30].

Theoretically, for highly disordered 3D systems, a τ_{ϕ} possessing a very weak T dependence in a certain temperature interval and then crossing over to a slow increase with decreasing T has recently been predicted in a model based on tunneling states of dynamical structural defects [4]. This model also predicts a “counterintuitive” scaling $\tau_{\phi} \propto \rho$ in the plateaulike region. Our observations in Fig. 2 essentially mimic these qualitative features. How this theory may be generalized to the 2D case should be of great interest. Experimentally, a scaling $\tau_{\phi} \propto D^{-1} \propto \rho$ for a good number of 3D polycrystalline alloys has recently been reported by Lin and Kao [19]. A dephasing time τ_{ϕ} (10 K) increasing with disorder $(k_F l)^{-1}$ in 3D $\text{In}_2\text{O}_{3-x}$ thin films [16] and a τ_{ϕ} (4.2 K) $\propto \rho$ in 2D $\text{In}_2\text{O}_{3-x}$ thin films [15] have also been found. Those results and Fig. 2 may indicate some generality of the dynamic defects in causing dephasing at low T [2–5]. This, yet-to-be fully understood, very strong and nonmagnetic dephasing mechanism deserves further investigation.

We report the observation of a strong electron dephasing in disordered $\text{Cu}_{93}\text{Ge}_4\text{Au}_3$ thin films. This dephasing is much stronger than any known inelastic electron scattering process. Our observation of a strict $\ln T$ -dependent, but high-magnetic-field-insensitive, resistance rise indicates

that this dephasing must be nonmagnetic in origin. The recent theoretical concept [4,5] of the dynamic defects is qualitatively in line with our result. This work provides key information for uncovering the long-standing saturation problem of τ_{ϕ} in mesoscopic systems.

We acknowledge helpful discussions with C. Bäuerle, Y. M. Galperin, V. Vinokur, A. Zaikin, G. Zarand, and A. Zawadowski. This work was supported by the Taiwan NSC through Grant No. NSC 94-2112-M-009-035, by the MOE ATU Program, and by the RIKEN-NCTU Joint Graduate School Program (S. M. H.).

*jjlin@mail.nctu.edu.tw

- [1] D. S. Golubev and A. D. Zaikin, Phys. Rev. Lett. **81**, 1074 (1998).
- [2] A. Zawadowski *et al.*, Phys. Rev. Lett. **83**, 2632 (1999).
- [3] Y. Imry *et al.*, Europhys. Lett. **47**, 608 (1999).
- [4] Y. M. Galperin *et al.*, Phys. Rev. B **69**, 073102 (2004).
- [5] Y. Imry *et al.*, arXiv:cond-mat/0312135.
- [6] P. Mohanty *et al.*, Phys. Rev. Lett. **78**, 3366 (1997); P. Mohanty and R. A. Webb, *ibid.* **91**, 066604 (2003).
- [7] D. Natelson *et al.*, Phys. Rev. Lett. **86**, 1821 (2001).
- [8] F. Pierre and N. O. Birge, Phys. Rev. Lett. **89**, 206804 (2002); F. Pierre *et al.*, Phys. Rev. B **68**, 085413 (2003).
- [9] F. Schopfer *et al.*, Phys. Rev. Lett. **90**, 056801 (2003).
- [10] G. Zarand *et al.*, Phys. Rev. Lett. **93**, 107204 (2004).
- [11] T. Micklitz *et al.*, Phys. Rev. Lett. **96**, 226601 (2006); T. Micklitz *et al.*, Phys. Rev. B **75**, 054406 (2007).
- [12] S. Kettemann and E. R. Mucciolo, Phys. Rev. B **75**, 184407 (2007).
- [13] F. Mallet *et al.*, Phys. Rev. Lett. **97**, 226804 (2006).
- [14] G. M. Alzoubi and N. O. Birge, Phys. Rev. Lett. **97**, 226803 (2006).
- [15] Z. Ovadyahu, J. Phys. C **16**, L845 (1983).
- [16] Z. Ovadyahu, Phys. Rev. Lett. **52**, 569 (1984).
- [17] Z. Ovadyahu, Phys. Rev. B **63**, 235403 (2001).
- [18] J. J. Lin *et al.*, Europhys. Lett. **57**, 872 (2002); J. J. Lin and N. Giordano, Phys. Rev. B **35**, 1071 (1987); J. J. Lin *et al.*, J. Phys. Soc. Jpn., Suppl. A **72**, 7 (2003).
- [19] J. J. Lin and L. Y. Kao, J. Phys. Condens. Matter **13**, L119 (2001).
- [20] S. Hikami *et al.*, Prog. Theor. Phys. **63**, 707 (1980).
- [21] J. J. Lin and J. P. Bird, J. Phys. Condens. Matter **14**, R501 (2002). For our films, $\tau_{e\text{-ph}} \approx (0.53 \pm 0.1)T^{-2}$ ns K⁻².
- [22] B. L. Altshuler *et al.*, Sov. Sci. Rev., Sect. A **9**, 223 (1987).
- [23] For a typical film with $R_{\square} = 30 \Omega$, Eq. (2.36) of Ref. [22] predicts $\tau_{e\text{-e}} \approx 1.1T^{-1}$ ns K⁻¹.
- [24] C. Van Haesendonck *et al.*, Phys. Rev. Lett. **58**, 1968 (1987); R. P. Peters *et al.*, *ibid.* **58**, 1964 (1987).
- [25] C. Bäuerle *et al.*, Phys. Rev. Lett. **95**, 266805 (2005).
- [26] J. Vranken *et al.*, Phys. Rev. B **37**, 8502 (1988).
- [27] M. D. Daybell and W. A. Steyert, Phys. Rev. Lett. **18**, 398 (1967).
- [28] P. Monod, Phys. Rev. Lett. **19**, 1113 (1967).
- [29] T. Cichorek *et al.*, Phys. Rev. B **68**, 144411 (2003).
- [30] The dynamic defects might be associated with the doped Au or Ge atoms. However, the precise nature requires further studies.