Weak localization effects in granular metals

C. Biagini,¹ T. Caneva,² V. Tognetti,^{2,3,4} and A. A. Varlamov⁵

¹INFM, Unità di "Tor Vergata," Viale del Politecnico 1, 00133 Roma, Italy

²Dipartimento di Fisica, Via G. Sansone 1, 50019 Sesto F. no (Firenze), Italy

³INFN, Sezione di Firenze, Via G. Sansone 1, 50019 Sesto F. no (Firenze), Italy

⁴INFM, Unità di Firenze, Via G. Sansone 1, 50019 Sesto F. no (Firenze), Italy

⁵COHERENTIA-INFM, CNR, Viale del Politecnico 1, 00133, Roma, Italy

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The weak localization (WL) correction to the conductivity of a granular metal is calculated using the diagrammatic technique in the reciprocal grain lattice representation. The properties of this correction are very similar to the corresponding one in disordered metal, with the replacement of the electron mean free path ℓ by the grain diameter d and the dimensionless conductance g by the tunneling dimensionless conductance g_T . In particular, we demonstrate that at zero temperature no conducting phase can exist for dimensions $D \leq 2$. We also analyze the WL correction to magnetoconductivity in the weak field limit.

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Recently, the properties of granular materials have attracted special attention.^{1–4} The quantization of the electron spectrum in small grains requires one to revise the basic idea of the quasiparticle spectrum continuity, assumed in the description of most properties of metallic and superconducting systems. The appearance of a new energy scale, the mean level spacing δ , results in unusual superconducting properties of such systems, with a possibility of observing, in these systems, specific quantum phase transitions, etc. In particular, it turns out that the interplay between the intragrain diffusion and intergrain tunneling of electrons makes the metalinsulator transition in such "quantum metal" very peculiar. In this Communication, we intend to discuss the specifics of the weak localization corrections in granular systems.

The elastic electron relaxation rate in granular metal consists of three contributions

$$\frac{1}{\tau_{\rm el}} = \frac{1}{\tau_{\rm imp}} + E_T + \Gamma, \tag{1}$$

where τ_{imp} is the mean scattering time of electrons with impurities, $E_T = v_F/d$ is the Thouless energy, *d* is the characteristic grain size, and v_F is the intragrain Fermi velocity. The last term is the electron intergrain tunneling rate: $\Gamma \sim g_T \delta$, where $g_T = 2\pi (t/\delta)^2$ is the tunneling dimensionless conductance and *t* is the tunneling energy.

The character of electron motion in a metal is conveniently classified as a function of its mean free path ℓ .⁵ The diffusion length $\ell_T^{(n)} = \sqrt{D_n/T}$ (where $D_n = v_F^2 \tau_{imp}/D$ is the diffusion coefficient of metal and *D* is the space dimensionality) separates the regions of ballistic ($\ell \ge \ell_T$) and diffusive ($\ell \le \ell_T$) electron motion. When $\ell \rightarrow \hbar/p_F$ the metal-insulator transition in the three-dimensional (3D) case takes place.

In the case of a granular metal, one can expect that the standard WL theory, describing the precursor effects of this transition,^{6,7} has to be modified in two ways. First, the diffusion coefficient \mathcal{D} here, at least in some interval of parameters, has to be determined by the tunneling time Γ^{-1} instead of elastic scattering time τ_{imp} and, therefore, we will

have a different tunneling thermal length $\ell_T = \sqrt{D}/T$. Second, the characteristic grain size d must appear in the theory side by side with the diffusion length ℓ_T . Two different situations are possible. When $d \ll \ell_T$, the diffusive part of the ℓ axis is divided into the regions of the normal intra-grain diffusion $(\hbar/p_F \ll \ell \ll d)$ with the metallic diffusion coefficient \mathcal{D}_n and of the tunnel intergrain electron diffusion $(d \ll \ell \ll \ell_T)$ characterized by the diffusion coefficient $\mathcal{D}=(\langle \mathbf{x}(t)^2 \rangle / t)_{t\to\infty}$ $\sim \Gamma d^2$ [see Fig. 1(a)]. In the opposite limit, when $d \gg \ell_T$, only the intragrain diffusion is possible, but, in its turn, the domain of ballistic regime contains two regions: the intragrain one with ballistic time τ_{imp} ($\ell_T \ll \ell \ll d$) and the region of intergrain electron motion with the ballistic time Γ^{-1} [see Fig. 1(b)].

We will discuss here the most interesting case of low temperatures, $d \ll \ell_T$. We will also assume that the electron motion inside a single grain is ballistic $(\ell_{bulk} \ge d, \ell \ge d)$: this means that before tunneling to the neighbor grain the electron is reflected many times on the grain boundaries ($\Gamma \ll E_T$). As a consequence, $\tau_{el} \approx E_T^{-1}$ and the dimensionless conductance of a single grain $g = E_T / \delta \ge g_T$. The conductance of the entire system is given by $g^{-1} + g_T^{-1} \approx g_T^{-1}$, what is



FIG. 1. A schematic representation of the two possible scenarios for the electron motion in granular metals as a function of the ratio between *d*, the grain diameter, ℓ_T , the thermal length, and ℓ , the mean free path.

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equivalent to saying that the drop of applied electric potential occurs only inside the tunnel barrier.

We assume that the grains are almost identical, with average diameter d, and form a regular lattice with lattice constant equal to the same d. The coordinate of the grain center will be labeled by the lattice variable \mathbf{R}_i . The Hamiltonian of the system can be written as¹

$$\hat{H} = \sum_{i,\mathbf{p}} \varepsilon_{\mathbf{p}} \hat{c}_{i,\mathbf{p}}^{\dagger} \hat{c}_{i,\mathbf{p}} + \frac{1}{2} \sum_{\langle i,j \rangle} \sum_{\mathbf{p},\mathbf{p}'} [t_{ij}^{\mathbf{p},\mathbf{p}'} \hat{c}_{i,\mathbf{p}}^{\dagger} \hat{c}_{j,\mathbf{p}'} + \text{H.c.}],$$

where $\hat{c}_{i,\mathbf{p}}$ ($\hat{c}_{i,\mathbf{p}}^{\dagger}$) is the annihilation (creation) operator of an electron in grain *i* with intragrain momentum **p**. The tunneling energy $t_{ij}^{\mathbf{p},\mathbf{p}'}$ will be taken equal for all bonds between nearest-neighbor grains and independent on the intragrain momentum, $t_{ij}^{\mathbf{p},\mathbf{p}'} = t$. Performing the Fourier transform with respect to such \mathbf{R}_i one can write the Hamiltonian in the representation of both intragrain and intergrain momenta (double momentum representation):

$$\begin{aligned} \hat{H} &= \sum_{\mathbf{K},\mathbf{p}} \left[\varepsilon_{\mathbf{p}} + tZ\gamma_{\mathbf{K}} \right] \hat{c}_{\mathbf{K},\mathbf{p}}^{\dagger} \hat{c}_{\mathbf{K},\mathbf{p}} \\ &+ \frac{tZ}{2} \sum_{\mathbf{K}} \sum_{\mathbf{p} \neq \mathbf{p}'} \gamma_{\mathbf{K}} \left[\hat{c}_{\mathbf{K},\mathbf{p}}^{\dagger} \hat{c}_{\mathbf{K},\mathbf{p}'} + \text{H.c.} \right]. \end{aligned}$$
(2)

Here **K** is the quasimomentum belonging to the reciprocal **R**_i lattice (it varies in the first Brillouin zone). The lattice structure factor $\gamma_{\mathbf{K}} = Z^{-1} \sum_{\mu=1}^{Z} e^{i\mathbf{K} \cdot \mathbf{d}_{\mu}}$; \mathbf{d}_{μ} are the vectors connecting the center of selected grain with the nearest-neighbor sites, *Z* is the coordination number. For a simple cubic lattice, the vectors \mathbf{d}_{μ} have one component equal to $\pm d$ and all the others equal to zero. For the sake of simplicity, we will restrict ourselves to this case; the extension to generic lattices is straightforward. In simple cubic lattices, $\gamma_{\mathbf{K}}$ $=1/D \sum_{\alpha=1}^{D} \cos(K_{\alpha}d)$. From Eq. (2) we can define the single electron Green function in the double momentum representation as

$$G_{\mathbf{K}}(\mathbf{p},\varepsilon_n) = \frac{1}{i\widetilde{\varepsilon}_n - \xi_{\mathbf{p}} - Zt(1 - \gamma_{\mathbf{K}})},$$
(3)

with $\tilde{\varepsilon}_n = \varepsilon_n + (2\tau_{el})^{-1} \operatorname{sign} \varepsilon_n$ and $\varepsilon_n = \pi T (2n+1)$ as fermionic Matsubara frequency.

Recalling that the electric field is negligible inside the grains and differs from zero only inside the barriers, in the presence of the vector potential **A** one can write the α th component of the electrical current operator in the imaginary time τ as

$$\hat{J}_{\alpha}(\tau) = i \frac{etd}{2} \sum_{\mathbf{K},\mathbf{p},\mathbf{p}'} \left[e^{iK_{\alpha}d} \hat{c}^{\dagger}_{\mathbf{K},\mathbf{p}}(\tau) \hat{c}_{\mathbf{K},\mathbf{p}'}(\tau) - \text{H.c.} \right] - \frac{e^2d}{2} [\mathbf{A}(\tau) \cdot \mathbf{d}] \hat{H}_T(\tau).$$
(4)

The linear response function, expressed as the second derivative of the partition function, is given by



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FIG. 2. Dyson equation for the diffusion vertex part in a granular metal in the direct space. The thick lines are the double Fourier transform of the fermionic Green functions Eq. (3), the dashed lines represent single impurity scattering and the initial and final grains are reported as roman indices i, j, etc. The Cooperon is obtained from the diffuson via time-reversal transformation on a fermionic Green function (Ref. 6).

$$\begin{aligned} \mathcal{K}_{\alpha,\alpha'}(\tau) &= -\frac{1}{\mathcal{Z}[0]} \left. \frac{\delta^2 \mathcal{Z}[A]}{\delta A_{\alpha}(\tau) \, \delta A_{\alpha'}(0)} \right|_{\mathbf{A} \to 0} \\ &= \Pi_{\alpha\alpha'}(\tau) - e^2 d^2 \delta_{\alpha\alpha'} \, \delta(\tau) \langle \hat{H}_T \rangle_0, \end{aligned} \tag{5}$$

where the current-current correlation function is expressed via the current operator as

$$\Pi_{\alpha,\alpha'}(\tau) = -\langle \hat{T}_{\tau} \hat{J}_{\alpha}(\tau) \hat{J}_{\alpha'}(0) \rangle_0.$$

The thermal average $\langle \rangle_0$ shall be performed with the diagonal Hamiltonian, the first line of Eq. (2). We are interested in the diagonal components of conductivity tensor:

$$\Pi_{\alpha,\alpha}(\omega_{\nu}) = 2e^{2}d^{2}|t|^{2}\sum_{\mathbf{K}} \sin^{2}(K_{\alpha}d)$$
$$\times T\sum_{\varepsilon_{n}}\sum_{\mathbf{p},\mathbf{p}'} G_{\mathbf{K}}(\mathbf{p},\varepsilon_{n+\nu})G_{\mathbf{K}}(\mathbf{p}',\varepsilon_{n}), \qquad (6)$$

where $\varepsilon_{n+\nu} = \varepsilon_n + \omega_{\nu}$ and $\omega_{\nu} = 2\pi T \nu$ is the bosonic Matsubara frequency. We can formulate the following rules of diagrammatic technique in the double momentum representation: (1) at each external vertex attach a factor \hat{v}_{α} $= etd \sin(K_{\alpha}d);$ (2) at each straight line attach a single electron Green function $G_{\mathbf{K}}(\mathbf{p}, \varepsilon_n)$; (3) sum over all internal momenta and Matsubara frequencies; (4) impose energy and lattice momentum conservation at each vertex. Now the impurity averaging of Eq. (6) can be performed. The Cooperon vertex⁶ corresponding to the granular metal can be obtained from the Dyson equation reported in Fig. 2. Another possibility is to renormalize the standard intragrain Cooperon by means of an introduction in the corresponding diagrams of a self-energy correction appearing due to tunneling, as it is done in Ref. 1. Both approaches in the assumptions made above turn out to be completely equivalent and lead to the expression

$$C_{\mathbf{Q}}(\omega_{\nu}) = \frac{1}{2\pi\tau_{\mathrm{el}}^{2}\nu_{F}} \frac{1}{|\omega_{\nu}| + 2\Gamma(1-\gamma_{\mathbf{Q}})}$$

In the latter expression the exact value appears for the tunneling rate: $\Gamma = Zg_T\delta$. In the expression $\mathcal{D} = [2\Gamma(1 - \gamma_{\mathbf{Q}})/|\mathbf{Q}|^2]_{\mathbf{Q}\to 0} = \Gamma d^2$ one can also recognize the effective "tunneling diffusion constant."

Now one can directly calculate the diagram reported in Fig. 3. In our assumptions, see Eq. (1), the Green function can be considered independent on **K** when integrating over



FIG. 3. WL correction to the conductivity in the double momentum representation. The solid lines are single electron temperature Green functions $G_{\mathbf{K}}(\mathbf{p},\varepsilon_n)$; the external velocity vertices are $\hat{v}_{\alpha} = etd \sin(K_{\alpha}d)$; the shaded box is the Cooperon $C_{\mathbf{O}}(\omega_{\nu})$.

p, because its behavior is completely determined by the pole due to the impurity (or grain boundaries) scattering, related to τ_{el}^{-1} , just as in the case of the diffusive limit for a bulk system. Performing the frequency summation, the **p** and **p'** integration and the sum over the lattice momentum **K** one finds

$$\frac{\mathcal{K}_{a\alpha}^{\mathrm{WL}}(\omega_{\nu})}{\omega_{\nu}} = -\frac{e^2 d^2}{4\pi\nu_F} g_T \sum_{\mathbf{Q}} \frac{\cos(Q_a d)}{|\omega_{\nu}| + 2\Gamma(1-\gamma_{\mathbf{Q}})}.$$

At this point, we can find the WL correction to the conductivity as

$$\frac{\delta\sigma_{(D)}^{WL}}{\sigma_{(D)}^{n}} = -\frac{1}{\pi Z g_T} \sum_{\mathbf{Q}} \frac{\cos(Q_{\alpha}d)}{1 - \gamma_{\mathbf{Q}}}.$$
(7)

In the case of a bulk granular system (D=3) the sum converges and the correction is finite. The metal-insulator phase transition can be observed at a critical value $Zg_T^{\text{cr}} \sim \mathcal{O}(1)$.

In the case of a granular film or wire $(D \le 2)$ the WL correction Eq. (7) diverges at small $\mathbf{Q} \to 0$. This fact indicates that at zero temperature these systems cannot exhibit metallic properties for any value of the dimensionless conductance g_T . At finite temperatures, a natural cutoff of Eq. (7) is provided by the phase-breaking rate $\gamma_{\varphi} = (\Gamma \tau_{\varphi})^{-1} = (d/L_{\varphi})^{2.6}$ It is related to the phase-coherence length $L_{\varphi} = \sqrt{\mathcal{D}\tau_{\varphi}}$, which tends to infinity when $T \to 0$. As a result

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$$\frac{\delta \sigma_{(1)}^{\text{WL}}}{\sigma_{(1)}^{n}} \approx -\frac{2}{Z\pi^{3}g_{T}}\frac{1}{\sqrt{\gamma_{\varphi}}};$$

$$\frac{\delta \sigma_{(2)}^{\text{WL}}}{\sigma_{(2)}^{n}} \approx -\frac{1}{Z\pi^{2}g_{T}}\ln\frac{\pi^{2}}{\gamma_{\varphi}}.$$
(8)

Equations (8) permit one to define the localization length $\xi_{(D)}^{\text{loc}}$ at which the correction becomes of the order of $\sigma_{(D)}^{n}$:

$$\frac{\xi_{(1)}^{\text{loc}}}{d} \approx \frac{Z\pi^3}{2}g_T;$$

$$\frac{\xi_{(2)}^{\text{loc}}}{d} \approx e^{Z\pi^2/2g_T}/\pi.$$

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It is worth noting that Eq. (7) does not contain τ_{el}^{-1} , but only lattice parameters as g_T and the coordination number Z: the intragrain dynamics, as expected, simply drops out of the calculation. The conductivity and its corrections are related to the diffusion on the grain lattice, and the mechanism of the momentum randomization between different grains is not crucial: the electron dynamics at low temperature can be thought of as that of a random walker on a lattice. This picture is fully consistent with the existing WL theory,^{6,7} and it is in agreement with previous experimental findings in granular metals.⁸

More intriguing are the properties of the WL correction to the magnetoresistance of granular metal. It is well known that quantum corrections to conductivity in disordered metal are very sensitive to the magnetic field: in fact, its presence disturbs the phase coherence of electrons moving along the self-intersecting trajectories, suppressing the WL correction and leading to the appearance of the anomalous negative magnetoresistance.⁶ In the following, we will show how such a correction manifests itself in the case of granular metal.

To calculate the WL contribution to magnetoresistance, it is necessary to rewrite Eq. (7) in the direct space:

$$\frac{\delta \sigma_{(D)}^{\text{WL}}}{\sigma_{(D)}^{n}} = -\frac{2}{Zg_{T}}\Gamma \frac{\widetilde{C}_{i,i+\alpha} + \widetilde{C}_{i+\alpha,i}}{2}$$

which is independent of *i*, depending only on the intergrain spacing *d*. α represents the bond along the direction of the current. Here $\tilde{C}_{\mathbf{Q}}^{-1} = (2\pi\tau_{\mathrm{el}}^2\nu_F C_{\mathbf{Q}})^{-1} = -i\omega + 2\Gamma(1-\gamma_{\mathbf{Q}})$. We notice that this form underlines the fact that transport is due only to the potential drop inside the barrier separating two grains *i* and *i*+ α . In the presence of a magnetic field, the Cooperon wave function is given by the solution of the equation

$$(4\Gamma)(1 - \gamma_{\mathbf{O}+2e\mathbf{A}})\psi_i(\mathbf{r}) = E\psi_i(\mathbf{r}).$$
(9)

Moreover, also the intragrain Cooperon will be renormalized by the presence of the magnetic field, acquiring a mass term equal to¹ $\mathcal{E}_0(H) = \frac{2}{5} (\pi \phi / \phi_0)^2 E_T$, where $\phi = Hd^2$ is the magnetic flux threaded through a single spherical grain and ϕ_0 $= \pi / e$ is the flux quantum. When the field satisfies the inequality $d \ll \ell_H = (eH)^{-1/2}$, or $\phi \ll \phi_0$, we have

$$\widetilde{C}_{ij}(\mathbf{r},\mathbf{r}',\omega) = \sum_{\mathcal{Q}_{\parallel},\mathcal{Q}_{\perp},n} \frac{\psi_{i,\mathcal{Q}_{\parallel}\mathcal{Q}_{\perp}n}(\mathbf{r})\psi_{j,\mathcal{Q}_{\parallel}\mathcal{Q}_{\perp}n}^{*}(\mathbf{r}')}{-i\omega + \Omega_{c}\left(n + \frac{1}{2}\right) + \mathcal{D}\mathcal{Q}_{\parallel}^{2} + \frac{1}{\tau_{\varphi}} + \mathcal{E}_{0}(H)},$$
(10)

where $\Omega_c = 4D/\ell_H^2$ is the Cooperon cyclotron energy. Q_{\parallel} is the momentum along the magnetic field and (n, Q_{\perp}) are the quantum numbers of the Landau basis.

The most interesting case is the two-dimensional geometry, with the magnetic field applied across the plane of the sample⁶ for which the WL correction is

$$\frac{\delta \sigma_{(2)}^{\text{WL}}}{\sigma_{(2)}^{n}} = -\frac{\cos\left(\frac{\Omega_{c}}{2\Gamma}\right)}{\pi Z g_{T}} \sum_{n=0}^{n_{\text{max}}} \frac{1}{n + \frac{1}{2} + \left(\gamma_{\varphi} + \frac{\mathcal{E}_{0}}{\Gamma}\right) \frac{\Gamma}{\Omega_{c}}}$$

 $\langle \alpha \rangle$

Since the sum over Landau levels is evidently divergent at the upper limit we introduced the cutoff parameter $n_{\text{max}} = \pi^2 \Gamma / \Omega_c \gg 1$, at which the cyclotron frequency becomes the order of the zone edge $n_{\text{max}} \Omega_c \sim \mathcal{D}(\pi/d)^2$. The final result reads

$$\frac{\delta \sigma_{(2)}^{\text{WL}}}{\sigma_{(2)}^n} = -\frac{1}{\pi Z g_T} \mathcal{F}(\phi, \gamma_{\varphi}),$$

where we express the magnetic field in terms of the magnetic flux

$$\mathcal{F}(\phi, \gamma_{\varphi}) = \cos\left(2\pi\frac{\phi}{\phi_{0}}\right) \left[\psi\left(\frac{\pi}{4}\frac{\phi_{0}}{\phi} + \frac{1}{2} + \frac{\pi}{10}\frac{\phi}{\phi_{0}}\frac{E_{T}}{\Gamma}\right) - \psi\left(\frac{\gamma_{\varphi}}{4\pi}\frac{\phi_{0}}{\phi} + \frac{1}{2} + \frac{\pi}{10}\frac{\phi}{\phi_{0}}\frac{E_{T}}{\Gamma}\right)\right],\tag{11}$$

where $\psi(x)$ is the digamma function. The WL correction to the magnetoresistance $\delta\sigma(H) = \delta\sigma^{WL}(H) - \delta\sigma^{WL}(0)$ is obtained as

$$\delta\sigma(H) = -\frac{\sigma_0}{\pi Z g_T} \bigg(\mathcal{F}(H, \gamma_{\varphi}) - \ln\frac{\pi^2}{\gamma_{\varphi}} \bigg).$$
(12)

One more energy scale shows up in Eq. (12) with respect to the bulk case, namely the Thouless energy. In the limit of very weak fields $\phi/\phi_0 \ll \sqrt{1/4E_T\tau_{\varphi}}$, this energy scale is not observable in the magnetoresistance: the leading singular correction reduces to

$$\frac{\delta\sigma(H)}{\sigma_0} \approx \frac{2\pi}{3} \frac{1}{Zg_T} \left(\frac{\phi}{\phi_0}\right)^2 \propto H^2$$

which corresponds to the anomalous magnetoresistance of the standard theory. The granular behavior deviates from the bulk one at fields such that $\phi/\phi_0 \sim \Gamma/E_T$ where the intra grain term starts to dominate in the second digamma function in $\mathcal{F}(H, \gamma_{\varphi})$: for $\sqrt{1/4E_T\tau_{\varphi}} \ll \phi/\phi_0 \ll \gamma_{\varphi}$, the magnetoresistance correction acquires the logarithmic form

$$\frac{\delta\sigma(H)}{\sigma_0} = \frac{2}{\pi Z g_T} \ln \left(\sqrt{\frac{E_T}{\Gamma}} \frac{\phi}{\phi_0} \right).$$

Larger fields are out of the range of our approximated approach, in which the intragrain Landau levels, with the spectrum $\omega_c(n+1/2)$ with $\omega_c = 4D_n/\ell_H^2$, start to significantly contribute to the Cooperon wave function. Let us notice finally that the ultraviolet cutoff $n_{\text{max}} \sim E_T/\omega_c = \Gamma/\Omega_c$ remains the same.

In summary, we have developed a diagrammatic technique in a double-momentum representation for transport in granular metals. Using this technique, the weak localization corrections to the conductivity arise in a natural way and an explicit calculation shows the same low-temperature behavior as in bulk metals, but with the diffusion constant D_n replaced by the effective tunneling diffusion constant D= Γd^2 and the mean free path ℓ by the average grain diameter d. Our result agrees with Eq. (13) of Ref. 9 in the $\mathbf{Q} \rightarrow 0$ limit; however, our technique underlines the presence of the grain lattice, represented by the cosine factor in Eq. (7), reminiscent of the lattice structure factor $\gamma_{\mathbf{Q}}$. We also give an estimate of the magnetoresistance correction for very weak fields.

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- ⁵ In a granular metal, ℓ coincides with the bulk mean free path ℓ_{bulk} up to the grain size *d*. If $\ell_{\text{bulk}} \gtrsim d$, the grain is in the ballistic limit. On the other hand, the electron will loose memory of its initial momentum state after a time of the order of τ_{el} : Eq. (1) with the assumption $\Gamma \ll \max\{1/\tau_{imp}, E_T\}$ implies that the elec-

tron relaxes its momentum only due to intragrain effects. We will define as a new mean free path just the new length scale, which appears only in finite systems, over which the electron looses completely the memory of its initial momentum state, $\ell \equiv v_F \tau_{el}$.

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