Giant Microwave Absorption in Metallic Grains: Relaxation Mechanism

F. Zhou,[1] B. Spivak,[1] N. Taniguchi,[2] and B. L. Altshuler[3,4]
[1]*Physics Department, University of Washington, Seattle, Washington 98195*
[2]*Department of Physical Electronics, Hiroshima University, Higashi-Hiroshima 739, Japan*
[3]*Department of Physics, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139*
[4]*NEC Research Institute, 4 Independence Way, Princeton, New Jersey 08540*
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We show that the low frequency microwave absorption of an ensemble of small metallic grains at low temperatures is dominated by a mesoscopic relaxation mechanism. Giant positive magnetoresistance and very strong temperature dependence of the microwave conductivity is predicted. [S0031-9007(96)01091-5]

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Microwave absorption in an ensemble of metallic grains has been investigated both theoretically and experimentally in many papers (see Refs. [1–4]). At high temperatures quantum effects can be neglected and the effective microwave conductivity at frequency ω is given by the classical Debye formula

$$\sigma_D = \frac{\omega^2}{\sigma_{\rm cl}}. \tag{1}$$

Here $\sigma_{\rm cl}$ is the classical Drude conductivity determined by elastic scattering. At low temperatures the quantum nature of electronic states in grains becomes essential and statistics of the electron levels determines the microwave absorption [1–4]. All quantum effects considered so far result in the microwave conductivity of the order or smaller than σ_D. In this Letter we discuss the mechanism of microwave absorption which can be much stronger than the classical one given by Eq. (1).

At sufficiently high radiation frequency ω absorption is dominated by the resonant mechanism, i.e., by direct transitions between electron levels [1,2], and the microwave conductivity is determined by the probability density $P(s)$ of energy spacing s between adjacent levels. This quantity is usually described by the Wigner-Dyson distribution. At $s \ll \Delta$, where Δ is the mean level spacing, the probability density behaves as [5] (see inset, Fig. 1)

$$P(s) = C_\beta s^\beta / \Delta^{\beta+1}. \tag{2}$$

The exponent β is determined by global symmetries of the system. For the orthogonal ensemble $\beta = 1$ and $C_\beta = \pi^2/6$. In the unitary case, $\beta = 2$ and $C_\beta = \pi^2/3$. Finally, the symplectic ensemble is characterized by $\beta = 4$ and $C_\beta = 16\pi^2/135$.

It is well known that absorption of radiation in two level systems is determined at low ω by a relaxation mechanism. Mandelstam and Leontovich proposed this mechanism for sound attenuation in gases with slow internal degrees of freedom [6]. For microwave absorption in doped semiconductors the relaxation mechanism was suggested by Pollak and Geballe [7].

The contribution of this mechanism to the microwave conductivity of metallic grains σ_R depends strongly on the relation between Δ and the level broadening Γ. For $\Gamma \gg \Delta$, the relaxation mechanism of absorption in granular metals was shown in Ref. [8] to determine under certain conditions both microwave conductivity and magnetoconductance of metallic grains. Here we consider the opposite limit $\Gamma \ll \Delta$ when energy levels are well resolved. It turns out that the microwave absorption σ_R as well as magnetoconductance are gigantic in this limit, i.e., σ_R exceeds both Eq. (1) and results of Ref. [8] by several orders of magnitude.

We now discuss the physical picture of the mechanism of relaxation absorption of applied electric field of frequency ω. In adiabatic approximation temporal energy levels $\epsilon_i(t)$ oscillate with the same frequency. Since the populations of energy levels follow adiabatically the motion of the levels themselves, electron distribution becomes nonequilibrium without any interlevel transitions. Relaxation of this nonequilibrium distribution due to inelastic processes leads to entropy production and therefore to absorption of energy of the external field.

For simplicity, we will restrict ourselves to the low temperature case when $T \ll \Delta$. (Later we will also discuss the situation at $\Gamma \ll \Delta < T$ qualitatively.) We also assume that $\omega \ll T$ (here and elsewhere we put $\hbar = 1$). Under these conditions the relaxation mechanism of absorption turns out to be determined by rare grains where the first excited level is separated from the ground state by small energy $s \ll T$. This implies that the ground and the first excited states form two level systems which are effectively isolated from the rest of the spectrum.

Population $N(t)$ of the first excited state with the energy $s(t)$ is governed by the equation

$$\frac{dN(t)}{dt} = -\frac{N(t) - N_0(s(t))}{\tau_\epsilon}. \tag{3}$$

Here $N_0(s(t)) = \{1 + \exp[s(t)/T]\}^{-1}$ is the adiabatic equilibrium population of the first excited level and τ_ϵ is

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the relaxation time of the level due to the electron-phonon interaction. Equation (3) is applicable provided $\omega \ll T$.

It is crucial for our discussion that the relaxation rate for a two level system vanishes with s. For a two level system embedded into a 3D insulating environment with phonons, $\tau_\epsilon \sim s^{-4}$ for $s \to 0$. So rapid divergence of $\tau_\epsilon(s)$ at small s leads, as we will see, to divergences in σ_R. To provide the proper cutoff we need to take into account processes that lead to a finite, though small, relaxation rate τ_m^{-1} at $s = 0$. We write $\tau_\epsilon(s)$ in a form

$$\frac{1}{\tau_\epsilon(s)} = \frac{1}{\tau_0}\left(\frac{s}{\Delta}\right)^4 + \frac{1}{\tau_m}, \tag{4}$$

where τ_0 and τ_m can be estimated as

$$\frac{1}{\tau_0} \sim \frac{\Delta^5 L^2 l T}{\Omega_D^2 v_s^3}, \qquad \frac{1}{\tau_m} = \frac{1}{\tau_0}\frac{1}{\tau_0 \Delta}\left(\frac{T}{\Delta}\right)^3, \tag{5}$$

Ω_D is the Debye frequency in the metal, v_s denotes the velocity of sound, L is the size of the sample, and l is the elastic mean free path. To interpret Eq. (5) we present the first term in Eq. (4) in the form $(s^3/\Omega_D^2)(\Delta/s)(kl)(kL)^2 T/s$, where $k = s/\hbar v_s$ is the wave number of a phonon with the energy s. The first factor here corresponds to the conventional expression for relaxation rate in clean bulk metal. We are considering a transition between two particular levels without summing over the final states (as is usual in the bulk case); this gives rise to the second factor. In the dirty case ($kl < 1$) the electron phonon relaxation time is known to be suppressed [9] (third factor). If the phonon wavelength exceeds the system size L as well, electron-phonon coupling in the dipole approximation is reduced, and the fourth factor appears. Finally, at $s < T$ the large population number of phonons leads to the last factor.

At small enough temperatures the main contribution to τ_m^{-1} comes from the two-phonon process: One phonon with an energy $\hbar\Omega \sim T$ is emitted and another phonon with the energy $\hbar\Omega - s$ is absorbed simultaneously to provide a transition between the two states separated by the energy $s \ll T$.

At higher temperatures, other mechanisms can contribute to τ_m^{-1} substantially, and $\tau_m(T)$ should be determined experimentally.

To evaluate σ_R we consider the power Q absorbed in a grain. Given the amplitude of the microwave electric field E and the volume of the grain V,

$$Q = \omega \int dt \frac{ds}{dt} N(t), \qquad \sigma_R = \frac{Q}{E^2 V}. \tag{6}$$

In the Ohmic approximation we obtain the Debye-type expression for the averaged microwave conductivity

$$\sigma_R = \left\langle \left(\frac{ds}{dE}\right)^2 \right\rangle \frac{1}{V} \int ds\, P(s) \frac{dN_0}{ds} \frac{\omega^2 \tau_\epsilon(s)}{1 + [\omega\tau_\epsilon(s)]^2}. \tag{7}$$

Here $\langle \cdots \rangle$ stands for the averaging over the random scattering potential. The random matrix elements were assumed to be statistically independent from the spectrum. This assumption can be verified by a straightforward calculation. We will present this calculation elsewhere.

Distribution $P(s)$ of the nearest neighbor spacing s is known to be determined by the global (spin and/or time reversal) symmetry of the system. σ_R, because of its dependence on $P(s)$ as seen in Eq. (7), should be very sensitive to weak magnetic field H and to spin-orbit scattering rate $1/\tau_{\rm so}$. Here we restrict ourselves only to three asymptotic regimes: (i) orthogonal, when $H = 0$ and $\tau_{\rm so} = \infty$, (ii) unitary, when the magnetic field is strong, and (iii) symplectic ($H = 0$, while $\tau_{\rm so}$ is short). We also discuss the particularly interesting case of the combination of strong spin-orbit scattering and a weak magnetic field. Apart from this exception we do not present here explicit formulae for the absorption in the crossover regimes. Here we will consider only linear absorption.

Substitution of Eq. (5) into Eq. (7) gives

$$\sigma_R = \frac{\omega^2 \tau_0}{4VT} \left\langle \left(\frac{ds}{dE}\right)^2 \right\rangle \phi\left(\frac{\Delta}{2T}, \omega\tau_m, \frac{\tau_0}{\tau_m}\right), \tag{8}$$

where

$$\phi(a,b,c) = \int_0^\infty \frac{(x^4 + c)x^\beta\, dx}{(x^4 + c)^2 + (bc)^2} \frac{1}{\cosh^2(ax)}. \tag{9}$$

For orthogonal ($\beta = 1$) and unitary ($\beta = 2$) ensembles of metallic grains ($\tau_{\rm so} s^* \ll 1$) the typical value of s determining the integral contribution in Eq. (7) is

$$s \sim s^* = \Delta(\max[\omega\tau_0, \tau_0/\tau_m])^{1/4}, \tag{10}$$

provided $s^* \ll T, \Delta$. In this limit, σ_R is

$$\sigma_R = \frac{\pi\omega^2\tau_0}{16VT} \left\langle \left(\frac{ds}{dE}\right)^2 \right\rangle \left(\frac{\tau_m}{\tau_0}\right)^{1/2\beta} f_\beta(\arctan(\omega\tau_m)), \tag{11}$$

with

$$f_\beta(k) = \mathrm{cosec}\left(\frac{\pi}{2\beta}\right)(\cos k)^{1/2\beta} \cos\left(\frac{k}{2\beta}\right). \tag{12}$$

The amplitude of electric field decays over Thomas-Fermi screening length $r_0 = \sqrt{D/4\pi\sigma_{\rm cl}} \ll L$ from its vacuum value E down to $E\omega/\sigma_{\rm cl} \ll E$. (Here D is the diffusion constant of electrons.) This small electric field in the bulk gives the main contribution to the classical Debye formula (1). On the contrary, the sensitivity (12) is determined by the vicinity of the surface, where the electric field is of the order of E. A calculation analogous to that in Ref. [8] gives

$$\left\langle \left(\frac{ds}{dE}\right)^2 \right\rangle \sim \frac{e^4 r_0^4}{\beta\sigma_{\rm cl} V}. \tag{13}$$

As a result, in the orthogonal case

$$\sigma_R^o = C\alpha\sigma_D \times \begin{cases} \left(\frac{\tau_m}{\tau_0}\right)^{\frac{1}{2}} & \omega\tau_m \ll 1 \\ \left(\frac{1}{2\omega\tau_0}\right)^{1/2} & \omega\tau_m \gg 1 \end{cases}, \tag{14}$$

while for the unitary ensemble,

$$\sigma_R^u = C\alpha\sigma_D \times \begin{cases} \left(\frac{\tau_m}{\tau_0}\right)^{\frac{1}{4}} & \omega\tau_m \ll 1 \\ 2\cos\left(\frac{\pi}{8}\right)\left(\frac{1}{\omega\tau_0}\right)^{1/2} & \omega\tau_m \gg 1 \end{cases}, \tag{15}$$

where τ_0 and τ_m are given by Eq. (5) and

$$\alpha = \pi\Delta^2\tau_0/372T \tag{16}$$

and C is a constant of the order of unity which depends on the geometry of the grain and on the direction of the electric field. If the grain is cubic and the microwave field is perpendicular to its face, then $C = 1$. We assume that $\Delta \gg T \gg \tau_0^{-1}$. Therefore in both the orthogonal and unitary cases σ_R is much larger than the classical Debye conductivity σ_D of Eq. (1).

In the absence of electron-electron Coulomb interaction, a similar approach is applicable even at $T > \Delta > \Gamma$ when many well resolved levels participate in the absorption. In this case $\sigma_R \sim \alpha'\sigma_D$ and $\alpha' = e^2 r_0^2 V^{-1}\tau_\epsilon(T)$. This matches the results [8] at $\Delta \sim \Gamma$.

As usual, one can drive an orthogonal system into a unitary one by applying magnetic field H. This leads to a giant negative magnetoconductance. Here we consider it only qualitatively and only at $\omega\tau_m \ll 1$. The field needed to reduce σ_R^o to σ_R^u is

$$H \sim H(s^*) = \left(\frac{s^*}{\Delta}\right)^{\frac{1}{2}} H_0\,, \tag{17}$$

where given the cross section of the grain A and its dimensionless (in units $e^2/\hbar$) conductance g, the characteristic field H_0 is

$$H_0 = \frac{\hbar c}{eAg^{\frac{1}{2}}}\,. \tag{18}$$

The field $H(s^*)$ reduces σ_R dramatically from σ_R^o to σ_R^u.

Now let the spin-orbit scattering rate τ_{so}^{-1} exceed Δ. In this symplectic case $P(s) \sim s^4/\Delta^5$ and the integral over s in Eq. (6) is determined by $s \sim T$. At low $\omega \ll \tau_\epsilon^{-1}$ (which means $s^* \ll T$) we have

$$\sigma_R^s = C\,\frac{64\pi^2}{45}\,\sigma_D\alpha\,\frac{T}{\Delta}\,, \qquad \text{if } \omega \ll (T/\Delta)^4\tau_0^{-1}\,. \tag{19}$$

At small frequencies σ_R^s turns out to be temperature independent and larger than σ_D by the factor $\alpha T/\Delta \sim \tau_0\Delta \gg 1$. At high frequencies the condition $s^* \ll T$ is violated. If $T < s^*$ [or $(T/\Delta)^4 < \omega\tau_0$], the characteristic energy separation s in Eq. (7) turns out to be of order T. As a result σ_R is ω-independent for all ensembles and

$$\sigma_R^{o,u,s} = \frac{CC_\beta}{4\pi\beta(\beta+5)}\,(T/\Delta)^{\beta+3}\,\frac{\sigma_{\rm cl}}{\tau_0}\,T\,. \tag{20}$$

This equation is valid as long as $\omega \ll T$.

In a sympletic grain each energy level is double degenerated due to the T invariance. An applied magnetic field splits this Kramers doublet and therefore increases $P(s)$ at small s. According to Kravtsov and Zirnbauer [10]

$$P(s,H) - P(s,0) \sim \frac{s^2}{s(H)^3}\,\exp\left[-\frac{s^2}{s(H)^2}\right]. \tag{21}$$

Therefore the distribution function has a peak at

$$s = s(H) \equiv \Delta\,\frac{H}{H_0} \tag{22}$$

and $P(s) \sim s^2$ below this peak. Calculation of the sensitivity $\langle(ds/dE)^2\rangle$ and τ_ϵ for a split Kramers doublet requires some caution: Without magnetic field both the external electric field and lattice deformation are unable to split a doublet (for example, $ds/dE = 0$ for $H = 0$). Therefore at $H < H_0$

$$\left\langle\left(\frac{ds}{dE}\right)^2\right\rangle \sim e^2 r_0^2\,\frac{\Delta}{4\pi\sigma_{\rm cl}}\left(\frac{H}{H_0}\right)^2, \tag{23}$$

$$\frac{1}{\tau_\epsilon} \sim \frac{1}{\tau_0}\left(\frac{s}{\Delta}\right)^4\left(\frac{H}{H_0}\right)^2 + \frac{1}{\tau_m}\,. \tag{24}$$

Substituting Eq. (21) into Eq. (7) we get the expression for grain magnetoconductance in the limit $\omega\tau_\epsilon \ll 1$ in the presence of substantial spin-orbit scattering. For $s(H) > s^*$, i.e., $H^4/H_0^4 > \omega\tau_0, \tau_0/\tau_m$, we get

$$\sigma_R(H) - \sigma_R(0) \sim \left(\frac{H_0}{H}\right)^3\sigma_R^u \gg \sigma^s(H=0)\,. \tag{25}$$

Thus we have a giant positive magnetoconductance that increases rapidly with the decreasing magnetic field until $H > H_0 s^*/\Delta$. Therefore at $H \sim H_0 s^*/\Delta$, the magnetoresistance has a sharp maximum. The magnetic field dependence of σ_R both for $\tau_{\rm so} \to \infty$ and for short $\tau_{\rm so}$ is qualitatively illustrated in Fig. 1.

Let us now estimate σ_R. Consider a metallic grain with $\Delta \sim 1$ K (the size is about $L \sim 150$ Å). τ_0 can be estimated as $\tau_0 \sim 10^{-7}$ sec. Then we get $\alpha \sim 10^2$ at $T \sim 0.3$ K. Therefore at $\omega \sim 10^5$ Hz we predict σ_R in the orthogonal case to be about 3 orders of magnitude larger than σ_D. Under these conditions $s^* \sim 0.3$ K and $H_0 \sim 10$ T (at $g \sim 10$), i.e., $H \sim 3$ T will make the absorption several times smaller.

Note that the magnetoconductance is negative and positive in the orthogonal and symplectic cases, respectively; i.e., the sign of the magnetoconductance is opposite to usual weak localization [11,12]. This is natural since classical microwave conductivity (1) is inversely proportional to $\sigma_{\rm cl}$.

So far we have neglected effects of the Coulomb interaction between electrons. First, our consideration was based on the Wigner-Dyson distribution $P(s)$. Its applicability in the presence of interactions is not obvious. Experimental study of microwave absorption could provide information on this probability distribution.

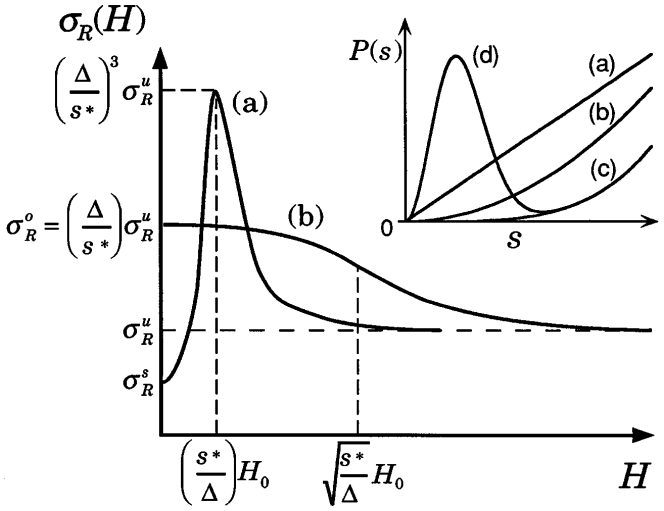

FIG. 1. Magnetic field dependence of the microwave conductivity at (a) strong spin-orbit scattering and (b) $\tau_{so} \to \infty$. Note a sharp and high peak in the first case due to the splitting of the Kramers doublets. Parameters $\sigma_R^{o,u,s}$, s^*, Δ, and H_0 are determined in the text. Inset: $P(s)$ at small s ($s \ll \Delta$): (a) orthogonal, (b) unitary, (c) symplectic, and (d) $\tau_{so}\Delta > 1$ and finite H.

Another Coulomb effect to consider is the contribution of the electron-electron interaction to the level broadening Γ. We argue that in the limit $T \ll \Delta$ this contribution does not exist and Γ is determined entirely by phonon scattering $\Gamma = \tau_\epsilon^{-1}$. Coulomb interactions can only renormalize the one-electron energy of the first excited state but do not broaden it because inelastic transitions due to electron-electron interaction are forbidden by the energy conservation law. Therefore Eq. (7) remains valid even for strongly correlated electrons.

As mentioned above, without Coulomb interaction there is a big interval $\Gamma \sim \tau_\epsilon^{-1} < \Delta < T$ where our two level approximation is not valid and many levels in each grain contribute to the absorption. These levels are still well resolved. On the other hand the electron-electron interaction is known to dominate Γ at high temperatures [12]. This makes calculations for $\Delta < T < \Delta(\tau_0\Delta)^{1/5}$ anything but trivial. Indeed, calculated in Ref. [12] Γ_{ee} is the broadening of one-particle states. The exact many-electron states can be broadened only by phonons. Spacing between these exact states $\Delta_{ex}(\epsilon)$ is of the order of Δ only for several low energy excitations. With increasing energy ϵ the spacing $\Delta_{ex}(\epsilon)$ decreases very quickly. We find it possible that the results of [8] are valid until $T > T^*$ where T^* is determined by the relation $\Delta_{ex}(T^*) = \Gamma_{ph}(T^*)$. If so, the matching should take place in the interval $\Delta < T < T^*$. We have to admit that much better understanding of the relation between exact states and quasiparticles and of electron-phonon interaction in the closed system is needed in order to develop a theory of microwave absorption at $\Delta < T < \Delta(\tau_0\Delta)^{1/5}$.

In addition to absorption of the electric field, one can consider the effect of an ac magnetic field. Here we mention only results of Ref. [13]. The authors of these papers assumed that the broadening of energy levels is energy independent and found that the energy dissipation in the time dependent magnetic field can be much larger than the classical one at $\Delta \gg \Gamma$. The sign of magnetoresistance in this case turns out to be the same as we found here, i.e., magnetoresistance is positive in an orthogonal ensemble and negative in a symplectic one. We believe, however, that the energy dependence of Γ is crucial for this problem as well, and we plan to present a quantitative theory elsewhere.

In conclusion, we showed that the relaxation microwave absorption at low frequencies and temperatures $\omega \ll T \ll \Delta$ turns out to be much stronger than the classical Debye mechanism [see Eqs. (14)–(16), (19), and(20)]. We also predict the giant and under certain conditions, nonmonotonic magnetoconductance.

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