Dephasing in Metals by Two-Level Systems in the 2-Channel Kondo Regime

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(Received 12 February 1999)

We point out a novel, nonuniversal contribution to the dephasing rate $1/\tau_\varphi \equiv \gamma_\varphi$ of conduction electrons in metallic systems: scattering off nonmagnetic two-level systems (TLSs) having almost degenerate Kondo ground states. In the regime $\Delta_{\rm ren} < T < T_K$ ($\Delta_{\rm ren}$ = renormalized level splitting, T_K = Kondo temperature), such TLSs exhibit non-Fermi-liquid physics that can cause γ_φ, which generally decreases with decreasing T, to seemingly saturate in a limited temperature range before vanishing for $T \to 0$. This could explain the saturation of dephasing recently observed in gold wires [Mohanty *et al.*, Phys. Rev. Lett. **78**, 3366 (1997)].

PACS numbers: 72.15.Qm, 72.10.–d, 72.70.+m, 73.50.–h

The dephasing behavior of conduction electrons in disordered systems in the zero-temperature limit has recently been subject to considerable and controversial discussions. The standard theory of dephasing, in the context of weak localization [1], predicts that the dephasing rate $1/\tau_\varphi \equiv \gamma_\varphi$ (extracted from the magnetoresistance) vanishes for $T \to 0$, since the phase space for inelastic scattering (e.g., electron-phonon or electron-electron) vanishes as the electron energy approaches the Fermi energy. In recent experiments on pure gold wires, however, Mohanty, Jariwala, and Webb (MJW) [2] found that γ_φ saturated at a *finite* value for $T \lesssim 1$ K, though two common "extrinsic" sources of dephasing, namely, magnetic impurities and heating, were demonstrably absent. Pointing out a similar saturation in older data on various other 1D and 2D diffusive systems, MJW suggested [2] that the saturation could be due to a universal mechanism intrinsic to the sample, namely, "zero-point fluctuations of phase coherent electrons." Although this suggestion contradicts the standard view, Golubev and Zaikin [3] developed it into a detailed theory that was claimed to agree with numerous experiments. However, their theory was criticized, most strongly in [4], but also in [5,6]. In [4] it was suggested that MJW's elaborate shielding precautions were insufficient and that external microwave fields caused the saturation.

In this Letter, we reexamine another source of dephasing, nonuniversal but intrinsic to any metal sample with structural disorder (which is never completely absent), namely, *dynamical two-level systems* (TLSs), such as point defects associated with dislocations, interfaces, surfaces, or amorphous regions. TLSs were not considered as a source of dephasing in the above-mentioned debate (except very recently in [7]), since standard inelastic scattering off nondegenerate TLSs (assuming the standard uniform distribution of level splittings, as discussed below) gives $\gamma_\varphi \sim T$ [8,9], which vanishes at low T.

Here, however, we point out that another dephasing mechanism exists for TLSs in metals: these are known to act as nonmagnetic or orbital 2-channel Kondo (2CK) impurities that exhibit non-Fermi-liquid (NFL) behavior in the regime $\Delta_{\rm ren} < T < T_K$ ($\Delta_{\rm ren}$ = renormalized level splitting, T_K = Kondo temperature), and we argue below that *this NFL behavior includes dephasing;* in fact, it can cause an apparent saturation in the decrease of γ_φ with decreasing T (although γ_φ does tend to zero for $T \to 0$). This novel dephasing mechanism is *nonuniversal*, since the distribution of the material parameter T_K sets the energy scale, and since the density of TLSs depends on the history of the sample. Reasonable assumptions for the density of TLSs in Au lead to estimates for γ_φ in accord with the saturation behavior seen by MJW.

We start by noting that any dynamical impurity, i.e., one with internal degrees of freedom, can potentially dephase a conduction electron scattering off it: if in the process the impurity changes its state, the electron's "environment changes," and this, quite generally, causes dephasing [10]. (In contrast, static impurities cannot change their state and hence cannot cause dephasing.)

In this Letter, we focus on dynamical "spin 1/2" impurities with two states, denoted by $\Uparrow$ and $\Downarrow$, which scatter free conduction electrons according to the rather general interaction (specific examples are discussed below):

$$H_I = \sum_{\varepsilon\varepsilon'} \sum_{\alpha\alpha' j \mu} c^\dagger_{\varepsilon\alpha j} v^\mu_{\alpha\alpha'} c_{\varepsilon'\alpha' j} S_\mu \, . \tag{1}$$

The electrons are labeled by an energy ε, a "spin" index α that is not necessarily conserved and a "channel" index j that (by definition) is conserved; the S_μ ($\mu = x, y, z$) are spin-1/2 operators, with S_z eigenvalues $(\Uparrow, \Downarrow) = (\frac{1}{2}, -\frac{1}{2})$; the coupling v^z describes the difference in scattering potentials seen by electrons scattering from the $\Uparrow$ or $\Downarrow$ state without flipping it, and is often called a "screening"

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interaction, since it generates a (S_z-dependent) screening cloud around the impurity; and v^x, v^y describe scattering processes that "flip the spin" of the impurity.

Magnetic impurities.—As an illustrative example, let us briefly review dephasing for the 1-channel Kondo model, for which the channel index $j = 1$ may be dropped and $v^{\mu}_{\alpha\alpha'} = v\sigma^{\mu}_{\alpha\alpha'}$. Let $\gamma(T)$ be the scattering rate of an electron at the Fermi surface ($\varepsilon = 0$); it can be split up as $\gamma = \gamma_\varphi + \gamma_{\rm pot}$ into parts that do or do not cause dephasing, respectively ("pot" for "potential" scattering). Two kinds of processes contribute to γ_φ: (i) Spin-flip scattering, as explained above, and (ii) single-to-many-particle scattering (see inset of Fig. 1), since additional electron-hole pairs can carry off phase information. Figure 1(a) shows the generic temperature dependence of γ, $\gamma_{\rm pot}$, and γ_φ [11]: As T approaches T_K from above, all three rates increase logarithmically. As T is decreased past T_K, γ continues to increase monotonically, but crosses over to a $(1 - \text{const} \times T^2)$ behavior; in contrast, γ_φ decreases (this has been observed directly in the magnetoresistance of samples containing magnetic impurities [2,12]), since below T_K the formation of a Kondo singlet between the impurity and its screening cloud begins to suppress spin-flip scattering. For $T \ll T_K$ the singlet is inert (with spin-flip rate $\sim e^{-T/T_K}$), and other conduction electrons experience only potential scattering off it; they, hence, form a Fermi liquid, in which a weak residual interaction between electrons of opposite spins [Eq. (D5) of [13]] yields a dephasing rate $\gamma_\varphi \propto T^2/T_K^2$, which vanishes as $T \to 0$.

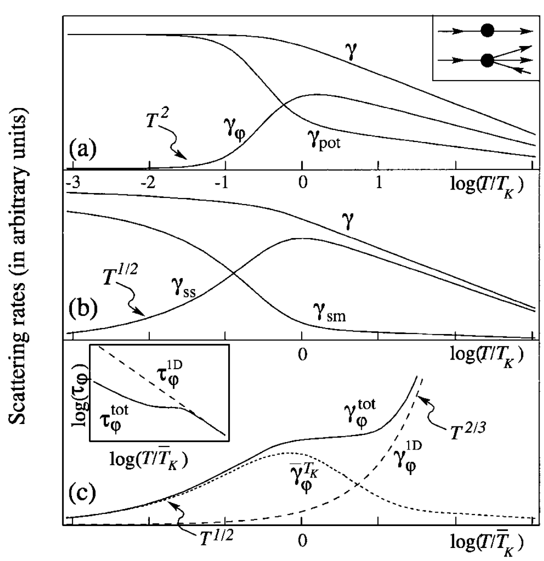

FIG. 1. Sketch of scattering rates as functions of $\log(T/T_K)$ for (a) the isotropic 1CK and (b) the anisotropic 2CK models (for $\Delta_{\rm ren} = 0$); inset: single-to-single-particle and single-to-many-particle scattering. (c) Dotted line: a Δ-averaged 2CK dephasing rate $\overline{\gamma}_\varphi^{T_K}$ with $T_K \simeq \overline{T}_K$ (averaging such curves over T_K yields $\overline{\gamma}_\varphi$); dashed line: $\gamma_\varphi^{\rm 1D} \sim T^{2/3}$; full line: $\gamma_\varphi^{\rm tot} = \overline{\gamma}_\varphi + \gamma_\varphi^{\rm 1D}$; inset: the corresponding dephasing times.

TLSs in metals.—Next we consider an atom or group of atoms moving in a double-well potential. Labeling the separate ground states of the "left" and "right" well by $(L, R) \equiv (\Uparrow, \Downarrow)$, the bare Hamiltonian $\Delta_z S_z + \Delta_x S_x$ describes a TLS with energy Δ_z, spontaneous transition rate Δ_x, and level splitting $\Delta = \sqrt{\Delta_z^2 + \Delta_x^2}$ between the ground and excited states, say, $|\pm\rangle$. It is common [9] to assume a constant distribution $P(\Delta) = \overline{P}$ of TLSs, with $\overline{P} \simeq 10^{19}$–$10^{20}$ eV^{-1} cm^{-3} in metallic glasses.

When put in a metal, such a TLS will scatter conduction electrons. The interaction's most general form is given by Eq. (1), where now the spin index α classifies the electron's orbital state, representing, e.g., its angular momentum (l, m), and the channel index $j = (\uparrow, \downarrow)$ denotes its Pauli spin, which is conserved since the TLS is nonmagnetic. This is in effect a generalized 2-channel Kondo interaction, with which one can associate a Kondo temperature T_K. In general, it is highly anisotropic, with $|v^x|, |v^y| \ll |v^z|$, since v^x, v^y describe electron-assisted interwell transitions and depend on the barrier size much more strongly than the screening interaction v^z does.

Slow fluctuators.—If the barrier is sufficiently large ($|v^x|, |v^y| \lll |v^z|$, so that $T_K \ll T, \Delta_x, \Delta_z$ and Kondo physics is not important), the system is a "slow fluctuator," which can adequately be described by the so-called "commutative" model, in which one sets $v^x = v^y = 0$ from the outset [9,14]. This model does not renormalize to strong coupling at low temperatures, and Δ is renormalized downward only slightly (by at most a few %) [15]. To estimate γ_φ, one may thus use the bare parameters and perturbation theory in v^z, which couples [9] $|+\rangle$ and $|-\rangle$. Since v_z scattering between these, being inelastic, requires $T > \Delta$, the Δ-averaged inelastic scattering rate is $\overline{\gamma}_{\rm inel} \propto T$ [provided that $(\Delta_x)_{\rm max}, (\Delta_z)_{\rm max} > T$ [8,9]]. Thus $\overline{\gamma}_\varphi \propto T$ also, which does not saturate as $T \to 0$.

Fast TLSs.—For sufficiently small interwell barriers, however, the effective T_K of a TLS can be significantly larger than its effective level splitting, so that Kondo physics does come into play [16]. Such TLSs require the use of the full "noncommutative" model with v^x and $v^y \neq 0$, which flows toward strong coupling under the renormalization group (RG) [17,18]. Extensive RG studies [19,20] showed that the regime $T \lesssim T_K$ is governed by an effective isotropic 2CK interaction of the form (1) with $\alpha = (1, 2)$ (since all but the two most-strongly coupled orbital states decouple) and $v^{\mu}_{\alpha\alpha'} = v\sigma^{\mu}_{\alpha\alpha'}$, and with an effective renormalized splitting $\Delta_{\rm ren} = \Delta^2/T_K$. In the so-called *NFL regime* $\Delta_{\rm ren} < T < T_K$, the resulting effective 2CK model exhibits NFL behavior [13,21]. The zero-bias anomalies observed in recent years in nanoconstrictions made from a number of different materials, such as Cu [22], Ti [23], or metallic glasses [24,25], can be consistently explained by attributing them to fast TLSs in or near this 2CK NFL regime [26,27]. The Kondo temperatures of the relevant TLSs were deduced in [22–24] from the width of the zero-bias anomalies to be $T_K \gtrsim 1$ K, and in [25] the insensitivity of the anomalies to a high-frequency modulation of the bias voltage implied $T_K \gtrsim 2$ K.

2CK Dephasing.—Let us now consider dephasing due to fast TLS in the NFL regime, a matter that to our knowledge has not been addressed before. In the NFL regime the single-to-single-particle and single-to-many-particle scattering rates $\gamma_{\mathrm{ss}}(T)$ and $\gamma_{\mathrm{sm}}(T)$ (inset of Fig. 1) are known to respectively decrease and increase with decreasing T, in such a way that the full scattering matrix is unitary [28]. Now, our key point is that *single-to-many-particle scattering must cause dephasing*, so that we can take $\gamma_\varphi \simeq \gamma_{\mathrm{sm}}$. This immediately implies that the dephasing rate *increases with decreasing temperature* in the NFL regime, as indicated schematically in Fig. 1(b). In fact, for $\Delta_{\mathrm{ren}} = 0$, one actually has $\gamma_{\mathrm{ss}} \propto (T/T_K)^{1/2} \to 0$ as $T \to 0$ [28], implying that the dephasing rate ($= \gamma_{\mathrm{sm}}$) would be finite even at $T = 0$. To heuristically understand this result, recall that the NFL fixed point describes an overscreened impurity that has a nonzero ground state entropy of $\frac{1}{2}\ln 2$ [21] and cannot be viewed as an inert object (in contrast to the 1CK case, where the ground state singlet has entropy 0); intuitively speaking, it is the dynamics associated with this residual entropy that causes dephasing even at $T = 0$.

Generally, however, $\Delta_{\mathrm{ren}} \neq 0$; for $T < \Delta_{\mathrm{ren}} = \Delta^2/T_K$, FL behavior is restored [20] and γ_φ drops back to zero, so that we crudely take $\gamma_\varphi(T) \simeq \gamma_{\mathrm{sm}}(T) \times \theta(\sqrt{TT_K} - \Delta)$. Since NFL physics also requires $\Delta < T_K$, we estimate the Δ-averaged dephasing rate [with $P(\Delta) = \overline{P}$] as

$$\overline{\gamma}_\varphi^{T_K} \simeq \int_0^{T_K} d\Delta\, P(\Delta)\gamma_\varphi(T), \qquad (2)$$

which yields $\overline{\gamma}_\varphi^{T_K}(T) \simeq \overline{P}\gamma_{\mathrm{sm}}(T)\min[\sqrt{TT_K}, T_K]$. This has a broad peak around T_K [Fig. 1(c), dotted line]. To next average over T_K, we assume that the distribution $P(T_K)$ has a broad maximum near, say, $\overline{T}_K$. Then the peak of $\overline{\gamma}_\varphi^{T_K}(T)$ would be broadened for $\overline{\gamma}_\varphi(T) = \int dT_K\, P(T_K)\overline{\gamma}_\varphi^{T_K}(T)$ into a flattened region near $\overline{T}_K$. Adding to this a power-law decay due to other sources of dephasing, e.g., $\gamma_\varphi^{\mathrm{1D}} \propto T^{2/3}$, the usual result for disordered 1D wires [1], the total dephasing rate $\gamma_\varphi^{\mathrm{tot}} = \overline{\gamma}_\varphi + \gamma_\varphi^{\mathrm{1D}}$ would have a broad shoulder around $\overline{T}_K$, while vanishing for $T \to 0$ [Fig. 1(c), solid line]. Thus 2CK impurities can cause the total dephasing rate $\gamma_\varphi^{\mathrm{tot}}(T)$ to seemingly saturate in a limited temperature range.

Estimate of numbers.—The shape of $\gamma_\varphi^{\mathrm{tot}}(T)$ and the existence of the broad shoulder depend on $P(T_K)$, $\overline{T}_K$ and the relative weights of $\overline{\gamma}_\varphi$ and $\gamma_\varphi^{\mathrm{1D}}$. To predict these from first principles would be overly ambitious, since a microscopically reliable model for the TLSs and their couplings to electrons is not available. Instead, let us use MJW's data to infer what properties would be needed to attribute their saturation to 2CK dephasing, and check the inferred properties against other studies of TLSs.

The dephasing times in Au wires saturated at $\tau_\varphi \simeq 5$ to 0.5 ns below a crossover temperature of about $T^* \simeq 1$ K, which we associate with $\overline{T}_K$. We further assume the saturation to be dominated by TLSs with $\Delta_{\mathrm{ren}} < T^* < T_K$, i.e., with $\Delta < 1\ \mathrm{K} < T_K$. Such parameters are reasonable, since experiment [22–25] and theory [16] suggest that a sizable fraction of $\Delta < 1$ K TLSs indeed do also have $T_K > 1$. Let us estimate their required density. Impurities with dephasing cross-section σ_φ and density n_i yield a dephasing rate $\gamma_\varphi = v_F n_i \sigma_\varphi$. The density of strongly coupled fast TLSs, i.e., with $\sigma_\varphi \lesssim \sigma_{\mathrm{unit}}$ close to the unitarity limit $\sigma_{\mathrm{unit}} = 4\pi/k_F^2$ per electron species, would thus have to be of order $n_i = 1/(\tau_\varphi v_F \sigma_{\mathrm{unit}}) \gtrsim 2 \times (10^{15}\text{–}10^{16})\ \mathrm{cm}^{-3}$ (which is rather small: given the atomic density in Au of $6 \times 10^{22}\ \mathrm{cm}^{-3}$, n_i implies a TLS density of only 0.02–0.2 ppm [29]).

The estimated value for n_i is reasonable, too: in metallic glasses, the density of TLSs with splittings $\Delta < 1$ K is $\overline{P} \times 1\ \mathrm{K} \simeq 9 \times (10^{14} - 10^{15})\ \mathrm{cm}^{-3}$; in polycrystalline Au, which is often taken to have roughly the same density of TLSs as metallic glasses [30], it is probably somewhat larger, since (i) in polycrystals, which constitute a more symmetric environment than glasses, the TLS distribution is probably more heavily weighted for small splittings; (ii) in 1D wires, surface defects can increase the total density of TLSs; and (iii) the bare splittings Δ_z, Δ_x are renormalized downward during the flow toward the NFL regime [20]. The density of TLS in Au wires that can be expected to cause 2CK dephasing thus compares satisfactorily with n_i estimated above.

Possible checks.—We emphasize that the 2CK dephasing mechanism is nonuniversal: first, the energy scale is set by T_K, and second, whether a sample contains sufficiently many TLSs to cause appreciable dephasing depends on its history. Thus, if the TLSs can be modified or even removed, e.g., by thermal cycling or annealing [22], the dephasing behavior should change significantly or even disappear. Drawn wires containing more dislocations (which may act as TLSs) should show stronger 2CK dephasing than evaporated wires [31]. Actually, already in 1987, Lin and Giordano [32] found hints in Au-Pd films of a low-temperature dephasing mechanism that is "very sensitive to metallurgical properties." In semiconductors, however, TLSs are unlikely to exhibit 2CK dephasing, since the much smaller electron density implies much smaller couplings (for recent dephasing experiments on semiconductors, see [4]).

In summary, we have pointed out a new, nonuniversal mechanism by which two-level systems in metals, acting as 2CK impurities, can cause dephasing, namely, through an increased single-to-many-particle scattering rate in their non-Fermi-liquid regime. We estimate that the Au wires of MJW [2] contain sufficiently many TLSs to yield 2CK dephasing rates comparable to the saturation rates observed there. More generally, though, the 2CK dephasing mechanism could be used to diagnose 2CK non-Fermi-liquid behavior in other metals containing TLSs.

We thank I. Aleiner, B. Altshuler, V. Ambegaokar, Y. Imry, N. Giordano, P. Nozières, H. Kroha, M. Vavilov,

A. Zaikin, G. Zaránd, and W. Zwerger for discussions. A. Z. enjoyed the hospitality of the Meissner Institute and the LMU in Munich; he was supported by the Hungarian Grants No. OTKA 96TO21228 and No. 97TO24005 and the Humboldt Foundation, J. v. D. by the DFG through SFB195, and D. C. R. by the MRSEC program of the NSF DMR-9632275 and the Packard Foundation.

Note added.—Concurrent with this work, Imry, Fukuyama, and Schwab [7] proposed that $1/f$ noise from TLSs might produce essentially T-independent dephasing by a different mechanism (not involving any 2CK physics), if $(\Delta_x)_{\max}$ is assumed to be $\ll T$ even for $T \simeq 1$ K, rather than the more common assumption [8,9] that Δ_x has a larger range.

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