Spin injection dependent metamagnetic transition

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We define the metamagnetic phase transition of itinerant electrons controlled by the spin injection mechanism. The current flow between a ferromagnetic metal and a metamagnetic metal produces the non-equilibrium shift of chemical potential for spin up and spin down electrons that acts as an effective magnetic field driving the metamagnetic transition.

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INTRODUCTION AND MAIN RESULTS

The idea of the spin injection from ferromagnetic metal to paramagnetic metal was first proposed by Aronov [1]. In the spin injection process the charge current flow between the ferromagnetic and paramagnetic metals produces the non-equilibrium magnetization in the paramagnet. This magnetization is proportional to the induced chemical potentials difference of electrons with opposite spins [1] - the spin accumulation. Non-equilibrium spin imbalance due to injection was observed by Johnson and Silsbee [2]. The theory of spin injection was developed in details in many works [3–7] and well studied experimentally, see for a review [8, 9]. However, the degree of electron spin polarization is relatively small at standard spin injection from ferromagnetic to paramagnetic metal [10, 11]. In order to increase the non-equilibrium polarization it is interesting to look for the possibility of spin-injection based magnetic transition in metamagnetic metals. Here we consider the metamagnetic transition of itinerant electrons induced by the spin injection mechanism. Let us briefly describe the properties of the metamagnet [12, 13]. When the energy splitting of electrons with opposite spins is smaller than the characteristic energy scale of itinerant electrons, then magnetic part of the free energy density can be expanded in powers of magnetic fields $F(H, M) = aM^2 + bM^4 + cM^4 - MH$, where coefficients $a$, $b$, $c$ are determined by the energy dependence of the density of states at the Fermi level, $H$ is the external magnetic field and $M$ is the magnetization.

At $b < 0$ magnetic part of free energy $F(H = 0, M)$ might have extremum at nonzero $|M|$ as it is shown in Fig. 1 which schematically illustrates evolution of free energy with increasing magnetic field due to contribution of the term $-MH$. At small magnetic field the state with low magnetization has lower energy, while at magnetic field larger than so called metamagnetic field $H_m$ the metamagnetic state acquires lower energy and system undergoes to state with higher magnetization. Metamagnetic state is induced by external magnetic field through the first order phase transition [12, 13].

Metamagnetic transition of itinerant electrons might appear [12, 13] in strongly enhanced Pauli paramagnets when the Fermi level is close to peak in electron density of states. In this case Zeeman splitting increases the density of states and drives the ferromagnetic transition.

The chemical potentials difference of electrons with opposite spins is the analog of external magnetic field in the non-equilibrium case. Characteristic feature of this effective magnetic field $H^{\text{eff}}(x)$ is the spatial non-uniformity which results in the finite length of the metamagnetic state. In the ferromagnet - metamagnetic metal contact spin accumulation and therefore effective magnetic field is generated in the region of the order of spin relaxation length at the vicinity of contact with ferromagnet and at the domain wall between metamagnetic - paramagnetic states. We assume that the domain wall thickness is much smaller than the spin relaxation length. Schematically ferromagnetic metal - metamagnetic state contact is shown in Fig. 2. Metamagnetic state is located at $0 < x < d$. Metamagnetic phase emerges at electric currents such that the effective magnetic field is $H^{\text{eff}}(x = 0) \geq H_m$. If $d$ is of order or larger than the spin relaxation length then effective field $H^{\text{eff}}(x)$ can be estimated as a sum $H^{\text{eff}}(x) = H^{\text{eff}}_{F-m}(x) + H^{\text{eff}}_{m-p}(x)$ of
the fields due to spin accumulation at boundary \( x = 0 \)

\[
H_{F-m}^{\text{eff}}(x) = \frac{eJ}{g\mu_B} \frac{2R_F R_m}{R_F + R_m} [\Pi_F - \Pi_m] e^{-|x|/\ell_m}
\]

and effective field due to spin accumulation at domain wall \( x = d \)

\[
H_{m-p}^{\text{eff}}(x) = \frac{eJ}{g\mu_B} \frac{2R_m R_p}{R_m + R_p} \Pi_m e^{-(d-x)/\ell_m}
\]

This case is shown by the solid line in Fig. 3. In expressions (1) and (2) \( J \) is the current density, \( e \) is the electron charge, \( \mu_B = |e|\hbar/2mc \) is the Bohr magneton and \( g = 2 \) for electrons,

\[
\Pi_{F,m} = \frac{\sigma_{\uparrow,F,m} - \sigma_{\downarrow,F,m}}{\sigma_{\uparrow,F,m} + \sigma_{\downarrow,F,m}}
\]

is proportional to the current polarizations, where \( \sigma_\alpha = e^2 D_\alpha \nu_\alpha \) are the corresponding conductivities in the ferromagnetic, metamagnetic and paramagnetic states for electrons with spin \( \alpha \), \( D_\alpha \) is the diffusion coefficient, \( \nu_\alpha \) is the density of states at the Fermi level,

\[
R_{F,m} = \ell_{F,m} \frac{\sigma_{\uparrow,F,m} + \sigma_{\downarrow,F,m}}{4\sigma_{\uparrow,F,m}\sigma_{\downarrow,F,m}}, R_p = \frac{\ell_p}{\sigma_p}
\]

are the effective resistances and the spin relaxation lengths are defined as \( \ell = \sqrt{D\tau_s} \), where in each state \( D = (D_\uparrow \sigma_\downarrow + D_\downarrow \sigma_\uparrow)/\left(\sigma_\uparrow + \sigma_\downarrow\right) \) and \( \tau_s \) is spin relaxation time.

In case of small thickness of domain wall transition between metamagnetic and paramagnetic states takes place at \( x = d \) when

\[
H^{\text{eff}}(d) = H_m
\]

as shown in Fig. 3. Taking the sum of expressions (1) and (2) we estimate the corresponding length of the metamagnetic region at \( d \geq \ell_m \) as

\[
d \sim \ell_m \ln \left| \frac{R_F [\Pi_F - \Pi_m]}{R_p [\Pi_F - \Pi_m]} \right| / (R_m + R_F)
\]

At large electrical current when \( g\mu_B H_m/eJ2R_m \to R_p \Pi_m/(R_m + R_p) \), according to expression (6) the length of metamagnetic region \( d \to \infty \). Threshold current density dependence of \( d \) occurs because of spin accumulation generation at domain wall between metamagnetic and paramagnetic states. However, since the effective field in most part of the region is below \( H_m \), the energy of stationary metamagnetic state at large \( d \) becomes smaller than the energy of paramagnetic state. We propose that, system must undergo to paramagnetic state at large values of current density. More detailed discussion of the transition is given below.

**ELECTRICAL SPIN INJECTION**

Consider the spin injection process from the ferromagnetic metal to metamagnetic metal. We focus on the spin and charge transport in the presence of the spin-orbit coupling and the short-range exchange electron-
electron interactions. We assume the vector of the nonequilibrium magnetization in the metamagnetic metal to be parallel to the vector of the magnetization in the ferromagnet. The Green’s function in Keldysh technique in the matrix form

\[ \hat{G} = \begin{pmatrix} \hat{G}^R & \hat{G}^K \\ 0 & \hat{G}^A \end{pmatrix} \]

is given by retarded \( \hat{G}^R(x,x') \), advanced \( \hat{G}^A(x,x') \) and Keldysh function \( \hat{G}^K(x,x') \), where \( x = (r,t) \) denote position and time arguments, hat (') stands for the matrix in spin space. Further we will consider the stationary regime in which function \( \hat{G} \) satisfies the equation

\[
\begin{align*}
\left( \omega + \frac{1}{2m} \nabla_r^2 + \mu - U(r) - e\phi(r) \right)i - \hat{U}_{so}(r) + \hat{\varepsilon}(r)\hat{G}(r, r', \omega) = \delta(r - r')
\end{align*}
\]

where \( \phi(r) \) is the electrostatic potential, \( U(r) \) is the random potential of the impurities assumed to be Gaussian distributed with \( \langle U(r) \rangle = 0, \langle U(r)U(r') \rangle = 2\pi\nu\nu\delta(r−r') \), \( \tau \) is the mean free time and \( \nu = (\nu_1 + \nu_2)/2 \) is the density of states. Spin-orbit interactions of electrons with impurities is described by the potential \( \hat{U}_{so}(r) = i\gamma\delta(\nabla U(r) \times \nabla) \), where \( \gamma \) is the spin-orbit coupling constant and \( \delta \) is the Pauli matrix [13]. The contribution of the short-range electron-electron exchange interactions to the spin splitting is described by the term \( \hat{\varepsilon}(r) \)

\[ \varepsilon(\mathbf{r}) = \frac{i\lambda}{2} \int \frac{d\mathbf{p}d\omega}{(2\pi)^4} G_{\kappa\alpha}(\mathbf{r}, \mathbf{p}, \omega) \]

where \( \lambda \) is the electron coupling constant and it is convenient to apply the Fourier transformation with respect to the relative coordinates \( r_1 = \mathbf{r} - \mathbf{r}' \) as

\[ \hat{G}(\mathbf{R}, \mathbf{p}, \omega) = \int d^3r_1 \hat{G}(\mathbf{R} + r_1/2, \mathbf{R} - r_1/2)e^{-i\mathbf{p} \cdot \mathbf{r}_1} \]

in which \( \mathbf{R} = (\mathbf{r} + \mathbf{r}')/2 \). The retarded and advanced Green functions \( \hat{G}^R \) and \( \hat{G}^A \) averaged over disorder in the \( \mu\tau \gg 1 \) approximation are diagonal in spin and are given by

\[ \hat{G}^{R,A}(\mathbf{R}, \mathbf{p}, \omega) = \frac{1}{\omega - \xi_\mathbf{p} + \hat{\varepsilon}(\mathbf{R}) \pm i\gamma} \]  

where \( \xi_\mathbf{p} \) is the electron dispersion, \( 2\gamma_\alpha = \tau^{-1}_\alpha + (t^{-1}_\alpha - \tau^{-1}_\alpha)/2 \) and \( \tau^{-1}_ \) is the inverse scattering times due to disorder and spin-orbit interactions for the spin state \( \alpha \), \( t^{-1}_\alpha = 4/3\tau_{so} \) [14]. We assume that the momentum relaxation time \( \tau_{so} \) is smaller than the time \( t_{so} \) corresponding to the spin flip process. Let us note, that we are considering the metamagnet when the exchange energy is the coordinate dependent function. In deriving the equation for the function \( \hat{G}^K \) we assume the limit when the exchange energy is small compared to the Fermi energy \( |\varepsilon_\downarrow - \varepsilon_\uparrow|/\mu < 1 \). In this limit the equation for the function \( \hat{G}^K \) yields

\[ \mathbf{v}(\nabla \mathbf{R} + [\nabla R\varepsilon_\alpha + e\mathbf{E}][\partial_{\varepsilon_\alpha}]G^K_K = -\left( \frac{1}{\tau_\alpha} - \frac{1}{\tau_{so}} + \frac{1}{\tau_{so}} \right) G^K_\alpha + \left( \frac{F_\alpha}{\tau_\alpha - \frac{F_\alpha - F_{\alpha-}}{2\tau_{so}} \right) [G^R_A - G^A] \]

here \( \mathbf{E} = -\nabla \phi(\mathbf{r}) \) is the electric field and we denote the coordinate and frequency dependent function

\[ F_\alpha(\mathbf{R}, \omega) = \frac{i}{2\pi\nu_\alpha} \int \frac{d\mathbf{p}}{(2\pi)^3} G^K_\alpha (\mathbf{R}, \mathbf{p}, \omega) \]

In the diffusion approximation for the function \( F_\alpha(\mathbf{R}, \omega) \) one obtains the equation

\[ \nabla \sigma_\alpha \nabla F_\alpha(\mathbf{R}, \omega) = \frac{\nu_\alpha\nu_- \alpha}{2\nu} F_\alpha(\mathbf{R}, \omega) - F_{\alpha-}(\mathbf{R}, \omega) \]  

where \( \sigma_\alpha = e^2\nu_\alpha D_\alpha \) is the conductivity, \( D_\alpha = v^2\tau_\alpha/3 \) is the diffusion coefficient and the density of states are the space dependent functions.

Let us consider the system when functions in Eq. [5] depend on one spatial coordinate \( (x) \) only. Consider the interface between a ferromagnetic metal that occupies the region \( (x < 0) \) and a metamagnetic metal \( (x > 0) \). We assume that the lengths of the ferromagnetic and metamagnetic regions \( L/2 \) are much larger than the corresponding spin diffusion lengths. We also assume the external reservoirs of the sample at \( x = \pm L/2 \) to be in the spin equilibrium state. The electric field in the system is treated through the boundary conditions

\[ F_\alpha(-L/2, \omega) = f(\omega - eV/2) \]
\[ F_\alpha(L/2, \omega) = f(\omega + eV/2) \]

where \( f(\omega) = \tanh(\omega/2T) \) and \( V = E/L \) is the potential difference across the structure. The solution of the Eq. [6] is the continuous function at the interface \( x = 0 \)

\[ F_\alpha(0-, \omega) = F_\alpha(0+, \omega) \]  

while the derivatives satisfy

\[ \sigma_\alpha \frac{\partial F_\alpha(x, \omega)}{\partial x} \bigg|_{x=0-} = \sigma_\alpha \frac{\partial F_\alpha(x, \omega)}{\partial x} \bigg|_{x=0+} \]  

describing the continuity of the current density at the interface. The current density carried by spin up and spin down electrons is given as

\[ J_\alpha(x) = \frac{1}{2\varepsilon} \int \sigma_\alpha \frac{\partial F_\alpha(x, \omega)}{\partial x} d\omega \]

We solve Eq. [8] assuming the boundaries ferromagnet-paramagnet, ferromagnet-metamagnet and metamagnetc-paramagnet independently. This approximation is valid in the limit when the length of the metamagnet \( d > \ell_m \). Taking into account the length of the system to be larger
than the corresponding spin-diffusion lengths we solve the diffusion equation in the region $x > 0$ with boundary conditions and continuity conditions.

\[
F_{\uparrow\downarrow}^{p,m}(x,\omega) = f(\omega + eV/2) + A_{p,m}[(x - L/2) \pm 2\sigma_{\uparrow\downarrow}^{p,m}(\Pi_F - \Pi_{p,m})\frac{R_F^pR_p^{m}}{(R_F + R_p^m)}e^{-x/\ell_{p,m}}] (12)
\]

where $p, m$ denotes low and high magnetization regimes of the metamagnet and $F$ stands for the ferromagnet, coefficient

\[
A_{p,m} = \frac{2(\sigma_{\uparrow F} + \sigma_{\downarrow F})[f(\omega + eV/2) - f(\omega - eV/2)]}{(|\sigma_{\uparrow F} + \sigma_{\downarrow F}| + (\sigma_{\uparrow p,m} + \sigma_{\downarrow p,m})L)}
\]

is connected with the current density as

\[
J = J_\uparrow(x) + J_\downarrow(x) = \frac{1}{2e} \int [\sigma_{\uparrow p,m} + \sigma_{\downarrow p,m}]A_{p,m}d\omega
\]

The conductivity spin polarization and resistivities in the ferromagnet and metamagnet are defined by expressions (9) and (10). Note, that in the low magnetization regime of the metamagnet $\sigma_{\uparrow p} = \sigma_{\downarrow p} = \sigma_p/2$, $D_{\uparrow p} = D_{\downarrow p}$.

Solution (12) has to be supplemented by the local neutrality condition which self-consistently determines the electric potential in the sample. The spin injection process does not change concentration of electrons

\[
N = \frac{1}{2} \int [\nu_\uparrow F_\uparrow(x,\omega) + \nu_\downarrow F_\downarrow(x,\omega)]d\omega (13)
\]

**PARAMAGNETIC STATE**

Low magnetization state of the metamagnet can be studied by solving Eq. (5) assuming contact between ferromagnetic metal and paramagnetic metal at $x = 0$. Effective field due to spin accumulation is

\[
H_{p}^{\text{eff}}(x) = \frac{eJ}{g\mu_B} \frac{2R_FR_p}{R_F + R_p}\Pi_F e^{-x/\ell_F}
\]

and magnetization at $x > 0$ is

\[
M_p(x) = \frac{(g\mu_B)^2\nu_p}{1 - \lambda\nu_p}H_{p}^{\text{eff}}(x) (14)
\]

Effective magnetic field produced by the spin accumulation in the ferromagnetic metal at $x < 0$ is

\[
H_{F_p}^{\text{eff}}(x) = \frac{eJ}{g\mu_B} \frac{2R.FR_F}{R_F + R_p}\Pi_F e^{x/\ell_F}
\]

where expressions for resistances $R_F$ and $R_p$ are given by Eq. (11).

**METAMAGNETIC TRANSITION**

The self-consistency equation for the magnetization density $M(x)$ in the sample is defined as

\[
M(x) = g\mu_B [\varepsilon_\uparrow(x) - \varepsilon_\downarrow(x)]/\lambda = -\frac{g_\mu_B}{2} \int [\nu_\uparrow F_\uparrow(x,\omega) - \nu_\downarrow F_\downarrow(x,\omega)]d\omega (16)
\]

In the case of equilibrium metamagnetic metal, Eq. (16) has two solutions even without the external magnetic field, corresponding to two minima of free energy, see inset in Fig. (1). Transition between these solutions takes place when magnetic field is equal to $H_m$. One could verify that in linear on $V$ response spin dependent part of expression (12) enters Eqs. (13) as magnetic field.

The procedure of finding solutions is following. We assume that there is metamagnetic state in the system at $0 < x < d$. Then we solve Eq. (8) for the spin accumulation at two boundaries and self consistently determine the value of $d$ from Eq. (3).

To obtain Eq. (4) we need to consider transition in more detail. Near transition between metamagnetic and paramagnetic states we need to include the spatial derivatives of magnetization into consideration, so

\[
-K\frac{d^2}{dx^2} M + \frac{\delta F(H_{M}^{\text{eff}}(x), M)}{\delta M} = 0
\]

Here $K$ is positive constant. Let we have solution $M_w(x - d)$, describing transition between metamagnetic and paramagnetic states at point $x = d$ in uniform magnetic field. It is solution of equation

\[
-K\frac{d^2}{dx^2} M_w + \frac{\delta F(H_{M,w}^{\text{eff}}(x), M_w)}{\delta M_w} = 0
\]

Assuming small difference $H_{M}^{\text{eff}}(x) - H_m$ at $x \approx d$ and substituting $M = M_w(x - d) + dM$, we obtain

\[
-K\frac{d^2}{dx^2}\delta M + \frac{1}{2} \frac{\delta^2 F(H_{M,w}^{\text{eff}}(x), M_w)}{\delta M^2}\bigg|_{M=M_w} dM = H_{M}^{\text{eff}}(x) - H_m
\]

Solution of this equation exists if

\[
\int dx \Psi_0(x)(H_{M}^{\text{eff}}(x) - H_m) = 0 (17)
\]

where $\Psi_0(x)$ is eigenfunction, corresponding to zero $E_0 = 0$ mode of equation

\[
-K\frac{d^2}{dx^2} \Psi_k + \frac{1}{2} \frac{\delta^2 F(H_{M,w}^{\text{eff}}(x), M_w)}{\delta M^2}\bigg|_{M=M_w} \Psi_k = E_k \Psi_k
\]

$\Psi_0(x)$ has no zeros and is localized near $x = d$ in region of order of domain wall thickness. This mode describes small translation of $M$, so in the uniform field one has $E_0 = 0$. In the case when $\ell_{m,p}$ are much larger than domain wall thickness from condition (17) one obtains Eq. (3).
Metamagnetic state

Let us discuss different realizations of spin accumulation.

1. Consider the case when $\Pi_F - \Pi_m$ and $\Pi_m$ have the same sign. Effective fields of both contacts have same sign too. The estimation for the length of metamagnetic region $d$ in the limit $d \geq \ell_m$ is given by expression (6). $d$ diverges at some threshold electrical current density.

2. Let $\Pi_F - \Pi_m$ and $\Pi_m$ have opposite signs. Thus, effective fields of both contacts have different signs too. Analysis shows that solution of Eq.5 with finite $d$ exist at $|H_{F-m}^{\text{eff}}(0)| > |H_{m-p}(d)|$. With increasing electrical current density $d$ stays finite.

In metamagnetic region the magnetization is

$$M_m(x) = M_m^0 + \frac{(g\mu_B)^2}{1 - \lambda \nu_m} H_{m}^{\text{eff}}(x)$$

Here $M_m^0$ is the magnetization of metamagnetic state, calculated at zero magnetic field and $\nu_m = 2\nu_m \nu_m^f/(\nu_m + \nu_m^f)$. Spin accumulation appears also in the ferromagnetic metal at $x < 0$ as

$$H_{Fm}^{\text{eff}}(x) = \frac{eJ}{g\mu_B} \frac{2R_F R_m}{R_F + R_m}[\Pi_F - \Pi_m] e^{x/\ell_F}$$

here expressions for $R_F$ and $R_m$ are given by (1).

Free energy criterium

We propose that the realization of metamagnetic state must be energetically favorable over realization of the paramagnetic state. In the linear on the applied current regime the magnetic part of free energy in the case of paramagnetic state realization is

$$\delta F_{Fp} = -M_F \int_{-L/2}^0 H_{Fp}^{\text{eff}}(x) dx$$

where $M_F$ is the magnetization of ferromagnetic contact. In the case of metamagnetic state realization it is

$$\delta F_{Fmp} = -M_F \int_{-L/2}^0 H_{Fmp}^{\text{eff}}(x) dx - M_m^0 \int_0^d [H_{m}^{\text{eff}}(x) - H_m] dx + F_S$$

Effective magnetic fields in ferromagnetic region are given by expressions (15) and (18). $F_S$ is the energy, associated with domain wall and boundary $F - m$. While domain wall energy is positive, the sign of $F - m$ boundary energy depends on relative directions of magnetizations in the ferromagnet and metamagnet. Estimation of $F_S$ depends on details that are beyond the scope of the paper.

From criterium $\delta F_{Fp} - \delta F_{Fmp} \geq 0$ for realization of the metamagnetic transition one can estimate the threshold value of current density. In the limiting case $R_F > R_m > R_p$, which assumes the contribution of the boundary $m - p$ and $F - p$ to the free energy is smaller than the corresponding contribution from the $F - m$ interface, one can estimate as

$$J_{thr} \approx \frac{g\mu_B}{e} \frac{H_m/2R_m}{(\Pi_F - \Pi_m)(1 + \ell_F M_F/\ell_m M_m^0)} d(J_{thr})$$

Ferromagnet-metamagnet-ferromagnet structure

Let us briefly discuss the spin injected metamagnetic state in system with metamagnetic metal placed between two ferromagnetic contacts with opposite directions of magnetizations. In this case $\delta F_{Fp} = 0$, because of cancellation of contributions in ferromagnets with opposite magnetizations. In metamagnetic state

$$\delta F_{FmF} = -M_m^0 \int_0^d [H_{m}^{\text{eff}}(x) - H_m] dx$$

Both ferromagnets contribute equally to the effective field. At $d \geq \ell_m$ using expression (11) we obtain the threshold value of electrical current density at which $\delta F_{FmF} \leq 0$ as

$$J_{thr} = \frac{g\mu_B}{e} \frac{H_m}{4R_F R_m |\Pi_F - \Pi_m|} \frac{R_F + R_m}{\ell_m} d$$

Note, that the expression for $J_{thr}$ for the $F - m$ contact in the limit discussed in the previous section is similar to the $F - m - F$ contact. Also note, that the transition to paramagnetic state with increasing current is absent.

CONCLUSIONS

To conclude, we have studied the metamagnetic transition of itinerant electrons in the metamagnet under the spin injection from the ferromagnetic metal. Spin injection produces the non-equilibrium effective magnetic field in metamagnet which drives the transition. We have calculated the effective magnetic fields and electrical currents required for the metamagnetic transition. We have shown that the length of metamagnetic state has threshold dependence on electrical current due to the effective magnetic field self generated at domain wall.

Typical values of the spin accumulation in metals are in the $\mu eV$ range [15, 16], which corresponds to the effective magnetic fields in tenth mT range at reasonably high current density. Metallic metamagnets with metamagnetic field in tesla’s range are well known [17]. Applying external magnetic field one can easily bring such system close to the transition.

Well studied YCo$_2$ in crystal form undergoes the metamagnetic transition at $H_m = 70T$ [13], while in the nanoscale structured form it is a weak ferromagnet [18],
suggesting the possibility of metamagnetic field reducing at proper technology. Other possibility is to study the system with temperature induced metamagnetic transition [19]. Unfortunately, spin relaxation length, the main parameter that governs the magnitude as well as the spatial distribution of effective field, is not known in such systems.

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