

## Generation of Spin Currents via Raman Scattering

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We show theoretically that stimulated spin-flip Raman scattering can be used to inject spin currents in doped semiconductors with spin-split bands. A pure spin current, where oppositely oriented spins move in opposite directions, can be injected in zinc blende crystals and structures. The calculated spin current should be detectable by pump-probe optical spectroscopy and anomalous Hall effect measurement.

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Spintronics is the strategy of using the spins of carriers in solid state structures as a new degree of freedom to carry and transform information [1]. An important capability is the generation of spin currents, which couple spin and charge. Suggestions for the generation of spin currents have included proposals based on the use of the extrinsic and intrinsic spin-Hall effects, as well as spin pumping [2–4]. All-optical generation and control of spin currents have also been proposed and observed [5–9]. In these optical schemes, the generation of the spin current is accompanied by the deposition of photon energies of about 1 eV per carrier in the system. Most of this energy is used simply to generate the electron-hole pairs and is lost in the sense that it does not contribute to the motion of the carriers. An obvious goal for the all-optical manipulation of spin is the minimization of such energy loss.

Here we propose a new scheme for generating spin currents in doped semiconductors via a two photon Raman scattering process [10,11], which can be understood as the absorption of a photon of frequency  $\omega_1$  and the emission of a photon at  $\omega_2$ . This process deposits an energy of  $\hbar\Omega = \hbar(\omega_1 - \omega_2)$  per carrier in the crystal, which for GaAs is only about 0.5 meV. In the presence of spin splitting of the bands, stimulated spin-flip Raman scattering can produce a pure spin current, where electrons with opposite spins move in opposite directions. Hence no net charge current is injected.

Crucial to this effect is the spin splitting of states, which results from the lack of inversion symmetry of the crystal. A schematic picture of the conduction bands is shown in Fig. 1, where  $E_F$  denotes the Fermi energy. For simplicity, we take the temperature to be zero [12]. The proposal in this Letter is based on optically inducing transitions between states  $d(\pm\mathbf{k})$  and  $u(\pm\mathbf{k})$  at  $\pm\mathbf{k}$  wave vectors, as depicted in Fig. 1, via Raman scattering. As an example, we first consider an optical field  $\mathbf{E} = \mathbf{E}(\omega_1)e^{-i\omega_1 t} + \mathbf{E}(\omega_2)e^{-i\omega_2 t} + \text{c.c.}$  in the material, where c.c. denotes complex conjugation and  $\omega_1 > \omega_2$ , and then present an experimentally relevant scenario of pulse excitation. The interaction of this field with the crystal in the dipole approximation is described by  $H_{\text{int}} = -(e/mc)\mathbf{p} \cdot \mathbf{A}(t)$ , where  $e$  ( $m$ ) is the charge (mass) of an electron,  $c$  is the

speed of light, and  $\mathbf{A}(t)$  is the vector potential associated with the field. In the ground state  $|0\rangle$ , electrons occupy states up to the Fermi level. The wave function of the perturbed system at time  $t$  is then written as  $|\Psi(t)\rangle = c_0(t)|0\rangle + \sum_{\mathbf{k}} c_{\mathbf{k}}(t)|e h \mathbf{k}\rangle$ , where  $|e h \mathbf{k}\rangle$  denotes the state where an electron ( $e$ ) and hole ( $h$ ) are generated at point  $\mathbf{k}$  (see Fig. 1). Time dependent perturbation theory is used to evaluate  $c_{\mathbf{k}}(t)$  and hence to describe the rate of change in the expectation value of a single particle operator  $\theta$  in the independent particle approximation,

$$\frac{\partial \langle \theta \rangle}{\partial t} = \sum_{\mathbf{k}} \langle e h \mathbf{k} | \theta | e h \mathbf{k} \rangle \frac{\partial [c_{\mathbf{k}}(t) c_{\mathbf{k}}^*(t)]}{\partial t}. \quad (1)$$

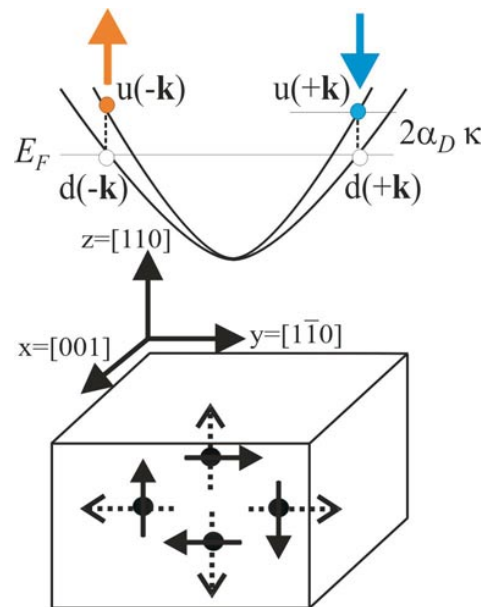


FIG. 1 (color online). Schematic picture of the spin-split conduction bands. Spin-flip Raman scatterings correspond to transitions from  $d(\pm\mathbf{k})$  to  $u(\pm\mathbf{k})$  at  $\pm\mathbf{k}$ . The carriers at  $u(\pm\mathbf{k})$  then have opposite spin orientations, and hence a pure spin current is injected in the system. An example of the generated spin current is presented where two pure spin currents with orthogonal velocities are shown schematically. The dotted lines correspond to the velocities of the carriers, and the thick arrows depict their spins.

This leads to the evaluation of injection rates of interest, including that for the density of flipped spins and for the spin current density [13]. The former can be written as

$$\frac{\partial n}{\partial t} = \xi^{abcd}(\omega_1, \Omega) E^a(\omega_1) E^{b*}(\omega_2) E^{c*}(\omega_1) E^d(\omega_2), \quad (2)$$

where  $\xi^{abcd}(\omega_1, \Omega) = V^{-1} \sum_{\mathbf{k}} \Gamma^{abcd}(\mathbf{k}) \delta(\Omega - \omega_{eh}(\mathbf{k}))$  and

$$\Gamma^{abcd}(\mathbf{k}) = \left( \frac{\sqrt{2\pi} e^2}{\hbar^2 \omega_2 \omega_1} \right)^2 \left\{ \left[ \sum_n \frac{v_{nh}^a(\mathbf{k}) v_{en}^b(\mathbf{k})}{\omega_1 + \omega_{hn}(\mathbf{k})} - \sum_n \frac{v_{en}^a(\mathbf{k}) v_{nh}^b(\mathbf{k})}{\omega_1 - \omega_{en}(\mathbf{k})} \right] \times (a \rightarrow c, b \rightarrow d)^* \right\}. \quad (3)$$

Here  $\omega_{nm} = \omega_n - \omega_m$ ,  $V$  is the normalization volume,  $a$ ,  $b$ ,  $c$ , and  $d$  are the Cartesian indices, and  $\mathbf{v}_{nm}(\mathbf{k})$  is the velocity matrix element between single particle states  $n$  and  $m$  at  $\mathbf{k}$ . Repeated indices are summed over. To quantify the spin current density  $J_{ab}$ , we use the conventional [14,15] single particle spin current operator  $j_{ab} = (1/2) \times (v^a S^b + S^b v^a)$ , where  $\mathbf{S} = \hbar \boldsymbol{\sigma} / 2$  and  $\boldsymbol{\sigma} = (\sigma^x, \sigma^y, \sigma^z)$  are the Pauli matrices. The spin current density injection rate is

$$\frac{\partial J_{ab}}{\partial t} = \mu^{abcdfg}(\omega_1, \Omega) E^c(\omega_1) E^{d*}(\omega_2) E^{f*}(\omega_1) E^g(\omega_2), \quad (4)$$

where  $\mu^{abcdfg}(\omega_1, \Omega) = V^{-1} \sum_{e,h,k} \delta j_{ab} \Gamma^{cdfg}(\mathbf{k}) \delta(\Omega - \omega_{eh}(\mathbf{k}))$  and  $\delta j_{ab} = \langle e\mathbf{k} | j_{ab} | e\mathbf{k} \rangle - \langle h\mathbf{k} | j_{ab} | h\mathbf{k} \rangle$  is the spin current contribution per spin flip [16]. Both the fourth rank tensor  $\xi^{abcd} = \xi^{cdab}$  and sixth rank pseudotensor  $\mu^{abcdfg} = \mu^{abfgcd}$  can be shown to be real by using time reversal symmetry; the  $T_d$  symmetry group of the crystal can be used to deduce the nonzero and independent tensor components. To calculate the tensors, we describe the spin splitting of the conduction states by the Dresselhaus Hamiltonian in the  $\Gamma$  point basis  $\{|1/2, 1/2\rangle, |1/2, -1/2\rangle\}$ :

$$H = \frac{\hbar^2 k^2}{2m_c} + \alpha_D \boldsymbol{\kappa} \cdot \boldsymbol{\sigma}, \quad (5)$$

where  $\kappa_x = k_x(k_y^2 - k_z^2)$  and  $\kappa_{y,z}$  are cyclic permutation of  $\kappa_x$ ,  $\alpha_D = 27.6 \text{ eV } \text{\AA}^3$  is the Dresselhaus parameter, and  $m_c = 0.067m$  is the electron effective mass [17,18]. The spin splitting at wave vector  $\mathbf{k}$  is  $2\alpha_D \kappa$ . The valence states of the semiconductor serve as the intermediate virtual states  $n$  in Eq. (3). The heavy and light valence states are modeled using the isotropic  $4 \times 4$  Luttinger-Kohn Hamiltonian, with the Luttinger parameters  $\gamma_1 = 6.85$  and  $\gamma = (\gamma_2 + \gamma_3)/2 = 2.5$ . The split-off band is described by a diagonal  $2 \times 2$  Hamiltonian, with the effective mass taken to be  $0.154m$  [19]. A band gap energy of  $E_g = 1.51 \text{ eV}$  and a split-off energy of  $0.34 \text{ eV}$  are used. The Fermi energy of the electrons in the conduction band is given by  $E_F = E_g + \hbar^2 k_F^2 / 2m_c$ , where  $k_F = (3\pi^2 N)^{1/3}$  for the carrier concentration  $N$ ; we use a concentration of  $N = 10^{18} \text{ cm}^{-3}$  in the calculations. The speed of the in-

jected carriers is given by  $v_F = \hbar k_F / m_c \approx 500 \text{ km/s}$ . The intraband velocity matrix elements are evaluated using  $\mathbf{v} = \hbar^{-1} \partial H(\mathbf{k}) / \partial \mathbf{k}$  as the velocity operator. We use a Kane parameter  $P = (\hbar/m) \langle S | p_x | X \rangle = 10.49 \text{ eV } \text{\AA}$  [19] and numerically evaluate the tensor components.

Spin orbit coupling splits the Fermi-surface into two surfaces. The fraction of the electrons that are contained in the region between the two surfaces is given by  $\approx 2m_c \alpha k_F / \hbar^2$  and is about 1% of the free carriers available in the system at  $N = 10^{18} \text{ cm}^{-3}$ . Only this fraction of the free carriers in the system is subject to Raman scattering. This sets a limit of  $\approx 10^{-2} N$  on the concentration of carriers that can contribute to the injected current.

Spin-flip Raman scattering has typically been studied using orthogonally polarized fields  $\mathbf{E}(\omega_{1,2})$  [11]. Hereafter, we use the following definition of the coordinates,  $\hat{x} \equiv [001]$ ,  $\hat{y} \equiv [1\bar{1}0]$ , and  $\hat{z} \equiv [110]$ , and consider more general fields normally incident on a crystal with a  $[110]$  growth axis

$$\mathbf{E}(\omega_{1,2}) = E_{1,2} \beta_{1,2} [\cos(\rho_{1,2}^x) \hat{x} + i \cos(\rho_{1,2}^y) \hat{y}], \quad (6)$$

for phases  $(\rho_1^x, \rho_1^y, \rho_2^x, \rho_2^y)$ , and normalization factors  $\beta_{1,2}$ . This allows the most general analysis of the fields polarizations. We find the spin flip and spin current density injection rates

$$\begin{aligned} \frac{\partial n}{\partial t} &= g A_{\varepsilon_1}(\Omega) I_1 I_2, \\ \frac{\partial J_{yz}}{\partial t} &= g \frac{\hbar}{2} [B_{\varepsilon_1}(\Omega) + C_{\varepsilon_1}(\Omega)] I_1 I_2, \\ \frac{\partial J_{zy}}{\partial t} &= g \frac{\hbar}{2} [B_{\varepsilon_1}(\Omega) - C_{\varepsilon_1}(\Omega)] I_1 I_2, \end{aligned} \quad (7)$$

where  $g = (\beta_1 \beta_2)^2 [\cos(\rho_1^y) \cos(\rho_2^x) + \cos(\rho_1^x) \cos(\rho_2^y)]^2$ , and  $I_{1,2}$  are the incident intensity of the  $\varepsilon_{1,2} = \hbar \omega_{1,2}$  components of the fields. Here  $A_{\varepsilon_1}$ ,  $B_{\varepsilon_1}$ , and  $C_{\varepsilon_1}$  are the linear combination of the tensor components  $\xi^{abcd}$  and  $\mu^{abcdfg}$ . The mismatch in the dielectric function  $\epsilon$  across the interface leads to a ratio  $2/(\sqrt{\epsilon} + 1)$  of the electric fields in the crystal and air. Two pure spin currents are injected in the crystal, one injected along  $y = [1\bar{1}0]$  with spins parallel to  $z = [110]$  and another injected along  $z = [110]$  with spins pointing parallel to  $y = [1\bar{1}0]$  (Fig. 1). Clearly, for either right (or left) circularly [e.g.,  $\beta_1 = \beta_2 = 1/\sqrt{2}$ ,  $\rho_1^x = \rho_2^x = 0$ ,  $\rho_1^y = \rho_2^y = 0$  (or  $\pi$ )] and cross linearly (e.g.,  $\beta_1 = \beta_2 = 1$ ,  $\rho_1^x = \rho_2^x = 0$ ,  $\rho_1^y = \rho_2^y = \pi/2$ ) polarized fields, the same spin currents are injected. Moreover, for colinearly polarized fields, where  $g = 0$ , no spin flips and hence no pure spin current can be induced.

First, we consider the case where the photon energy  $E_1 = \hbar \omega_1 = 0.754 \text{ eV}$  is close to  $E_g/2$ . All two photon absorption from the valence to the conduction bands is avoided in this scenario. In Fig. 2, we present  $A_1$ ,  $B_1$ , and  $C_1$ . The role of the resonance enhancement can be investigated by using optical fields of energy  $E_{\Pi} = \hbar \omega_1 = 1.49 \text{ eV}$  close to the

band gap. Transitions from the partially filled conduction band to the higher bands are avoided at this energy, but two photon excitations from the valence states to conduction states are allowed. Nonetheless, these transitions are away from the  $\Gamma$  point and should not contribute to the injected pure spin current from the spin-flip Raman processes we discuss in this Letter. The two photon absorption is estimated to be an order of magnitude smaller than the spin-flip injection rate discussed here [20], and excitons are not generated. The carrier and spin current density injection rates characterized by  $A_{II}$ ,  $B_{II}$ , and  $C_{II}$  are presented in Fig. 2. The sharp features in the results, shown in Fig. 2, correspond to the van Hove singularities in the joint density of states for the spin-flip transitions. The van Hove singularity is due to the vanishing derivative in the spin splitting,  $2\alpha_D \nabla_{\mathbf{k}} \kappa$  along high symmetry directions, and an analysis of the Dresselhaus Hamiltonian shows that the resulting density of states increases as  $\ln(1/|\Omega - \Omega_{vH}|)$ , where  $\Omega_{vH}$  is the position of the singularity. To this point, we have discussed the injection rate of pure spin currents by stimulated Raman scattering using two nominally continuous wave (CW) beams at distinct frequencies  $\omega_2$  and  $\omega_1 > \omega_2$ . We now turn to the ultrafast pulse regime, and stress that a pure spin current can be injected through the Raman effect by a single pulse, with the scattering now arising from the spectral width of the pulse. For a pulse with a

center frequency  $\omega_0$  and width  $\Delta\omega$ , if we have  $\omega_0 \gg \Delta\omega$  and  $\omega_0 \gg \Omega_{\max}$ , where  $\hbar\Omega_{\max}$  is the maximum spin splitting, the straightforward generalization of the calculation in Eqs. (2) and (4) leads to the injection of densities of spin flips and spin current given by

$$\Delta n = \int_0^{\Omega_{\max}} \frac{d\Omega}{2\pi} \xi^{abcd}(\omega_0, \Omega) G^{abcd}(\Omega), \quad (8)$$

$$\Delta J_{ab} = \int_0^{\Omega_{\max}} \frac{d\Omega}{2\pi} \mu^{abcdfg}(\omega_0, \Omega) G^{cdfg}(\Omega), \quad (9)$$

where the field correlation function for  $\mathbf{E}(t) = (2\pi)^{-1} \times \int_{-\infty}^{\infty} d\omega \mathbf{E}(\omega) e^{-i\omega t}$  is

$$G^{abcd}(\Omega) = \int_0^{\infty} \frac{d\omega_1 d\omega'_1}{(2\pi)^2} E^a(\omega_1) E^{b*}(\omega_1 - \Omega) E^{c*}(\omega'_1) E^d(\omega'_1 - \Omega). \quad (10)$$

We consider a (right or left) circularly polarized short pulse of the form  $E(t) = 2E_0 \exp(-t^2/\tau^2) \cos(\omega_0 t)$ , with fluence (pulse energy per unit area)  $F$ . A short pulse [21] ( $\tau\Omega_{\max} \ll 1$  and  $\tau \ll \mu m_c/|e|$ , where  $\mu$  is the mobility) essentially leads to the generation of the spin current in the ballistic limit. For  $\hbar\omega_0 = 0.754$  eV (corresponding to  $A_I$ ,  $B_I$ , and  $C_I$  in Fig. 2), the density of spin flips per pulse is  $\Delta n_I \approx (1.8 \times 10^{20} \text{ cm}^2/\text{J}^2) F^2$ , which gives  $\Delta n_I \approx 10^{15} \text{ cm}^{-3}$  for an experimentally accessible fluence of  $F = 3 \times 10^{-3} \text{ J}/\text{cm}^2$ . In a scenario where  $\hbar\omega_0 = 1.49$  eV (corresponding to  $A_{II}$ ,  $B_{II}$ , and  $C_{II}$  in Fig. 2), the spin-flip density due to Raman scattering is  $\Delta n_{II} \approx (1.2 \times 10^{22} \text{ cm}^2/\text{J}^2) F^2$ ; a spin-flip density of  $\Delta n_{II} \approx 10^{15} \text{ cm}^{-3}$  can be achieved for a fluence of  $F = 3 \times 10^{-4} \text{ J}/\text{cm}^2$ , which reflects a resonance enhancement of the process by 2 orders of magnitude.

Note that the injection in this process is proportional to  $F^2$ , whereas from Eq. (7) for nominal CW irradiation one might expect a scaling with  $I_{\max}^2 \tau \propto F^2/\tau$ , where  $I_{\max}$  is the maximum intensity in the pulse. The reason is that, for  $\Omega_{\max} \tau \ll 1$ , a factor  $\Omega_{\max} \tau \approx \Omega_{\max}/\Delta\omega$  appears in the final result following from Eqs. (8) and (9), reflecting the fact that only a fraction  $\Omega_{\max}/\Delta\omega$  of the pulse bandwidth can contribute to the Raman scattering.

With each pulse, a number of carriers  $\approx \Delta n \times S\eta$  is excited, where  $S$  is the laser spot area and  $\eta$  is the penetration depth. In earlier experiments [5,6] involving interband transitions, a large number of carriers is available in the valence states, and typically  $\approx (10^{18} \text{ cm}^{-3})S\eta$ , where  $\eta \approx 1 \mu\text{m}$  carriers can contribute to the spin currents. On the other hand, in Raman injected spin currents the penetration depth is much larger than that for interband transitions, with a limit  $\eta' \approx 10^3 \mu\text{m}$  set by the Drude absorption due to free carriers. This allows a large  $\approx (10^{15} \text{ cm}^{-3})S\eta'$  number of spin flips in the sample, which will then contribute to the injected pure spin current.

Pump and probe schemes have been used to experimentally observe injected pure spin currents [6]. An experi-

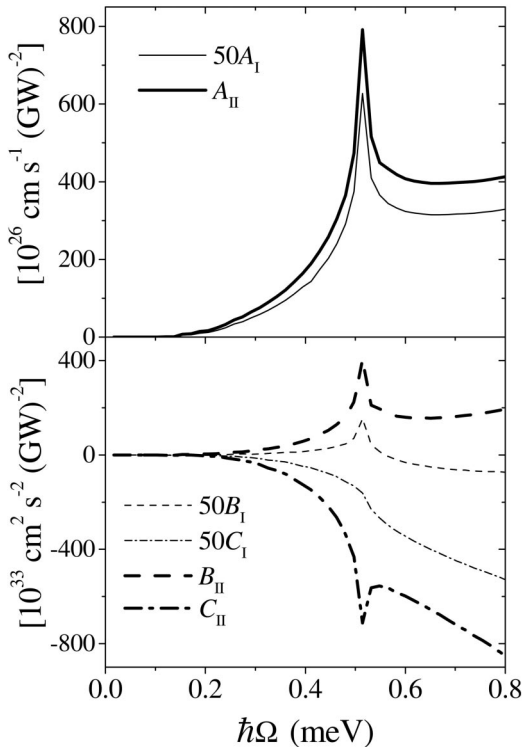


FIG. 2. The thin lines show the coefficients  $A_I$ ,  $B_I$ , and  $C_I$  Eq. (7) for  $\Omega = \omega_1 - \omega_2$  and  $\hbar\omega_1 = 0.754$  eV, about half the band gap. The thick lines show the coefficients  $A_{II}$ ,  $B_{II}$ , and  $C_{II}$  Eq. (7) for  $\hbar\omega_1 = 1.49$  eV, about 20 meV below the band gap.

mentally relevant parameter in these schemes is the separation between the opposite spins after momentum relaxation. This distance can be estimated by  $d^a = (2\mu m_c / |e\hbar|) \Sigma_b |\dot{J}_{ab}| / \dot{n}$  [22,23]. For circularly polarized as well as cross linearly polarized fields, a spin separation of  $d^y \approx 18$  nm for  $\mu \approx 5 \times 10^3$  cm<sup>2</sup>/(Vs) is predicted, a distance which is within experimental capabilities [6]. Measuring a pure spin current by using the anomalous spin-Hall effect arising due to skew scattering by impurities [24,25] has also been proposed [8]. The spin current  $J_{ab}$  causes a spin-Hall bias  $V_{\text{SH}}$  along the direction perpendicular to the electron propagation. This bias is estimated as  $V_{\text{SH}} \approx \tan(\theta_{\text{SH}}) V_{\text{eff}}$ , where  $\theta_{\text{SH}}$  is the spin-Hall angle and  $V_{\text{eff}}$  is the effective bias that would cause a current density  $eJ_{ab}/\hbar$ ; it is of the order of  $e\Delta n v_F$ , where  $\Delta n$  is the number of carriers that form the pure spin current. Therefore  $V_{\text{eff}} \approx L(\Delta n/N)v_F/\mu$ , where  $L \approx 10$   $\mu\text{m}$  is the lateral size of the system, which is assumed to be approximately equal to the spot size. The ratio  $\Delta n/N$  is estimated as  $\zeta \alpha_D k_F^3/E_F$ , where  $\zeta$  is the fraction of Raman accessible electrons that undergo a spin flip. With the above mobility, we obtain  $V_{\text{eff}}/L \approx 10\zeta$  V/cm. The above analysis shows  $\zeta$  to be about 0.1. With the angle  $\theta_{\text{SH}}$  estimated by Abakumov and Yassievich [24] for GaAs at low temperatures as  $\theta_{\text{SH}} \approx 10^{-3}$ , we obtain an upper estimate of  $V_{\text{SH}} \approx 10^{-6}$  V.

In conclusion, stimulated spin-flip Raman scattering can be used to inject spin currents in doped semiconductors with spin splitting. We have considered the case of an  $n$ -doped GaAs bulk crystal and have shown that the injected spin current is a pure spin current comprised of spins with opposite orientations traveling in opposite directions. This pure spin current should be experimentally accessible and can be achieved either with two continuous waves contributing to the Raman scattering or by using a single pulse. The scheme is all optical, but, unlike other optical schemes involving the excitation of electron-hole pairs, here there is minimal energy deposited in the crystal. The system is thus only slightly perturbed from equilibrium, and the description of transport and the design of device structures should be easier than in scenarios employing other optical schemes. High mobility quantum wells should provide ideal testing grounds for the general proposal discussed in this Letter.

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