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Intrinsic spin-Hall effect in aluminum

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Abstract – With the help of an artificially created spin imbalance that induces a charge imbalance unrelated to the Lorentz force, the relativistic spin-Hall effect (SHE) in 3D superpure monocrystalline aluminum was first studied. The nonlinear behavior of SHE in a magnetic field is revealed as a direct proof of the manifestation of the intrinsic (band) mechanism of the spin polarization of electrons in a nonmagnetic metal under spin-orbit interaction conditions with a minimized external impurity spin-polarization factor. Based on the data obtained, the spin-orbit coupling coefficient in aluminum is estimated.

Since the introduction of the idea of an additional degree of freedom of an electron called a spin and supported by the relativistic Dirac equation as an alternative to the Schrödinger equation, the behavior of the electron wave function resulting from it has revealed a number of new properties. In 1929, Mott first showed that one of them can be a chiral asymmetry of the scattering of electrons with different spins in the central force field of impurities under conditions of a relativistic spin-orbit (SO) bond [1]. After 40 years, Diakonov and Perel, based on this idea, predicted for nonmagnetic conductors the effect of curving trajectories of electrons with opposite spin orientation and their subsequent accumulation on opposite edges of the samples [2]. However, an increased interest in this effect, not so obvious for solids, arose only after the appearance of some experimental evidence of its manifestation [3,4]. Apparently, the most demonstrative result is an optical demonstration of the existence of SHE [5,6], which served as an impetus for active research of generation of spin currents and manipulation of spins. Since, however, the concept of the Mott impurity mechanism of asymmetric spin scattering was considered to be the most accepted, virtually all the experiments and the theoretical analysis of their results were carried out within the framework that satisfied this concept. Confirmation of this can be found, for example, in the review [7], where an almost exhaustive list of existing principal experimental works on SHE is given. At the same time, it was shown that a relativistic SO field, which removes two-fold spin degeneracy [8], in the presence of an external electric field can induce in the crystal a transverse spin-dependent electron velocity even in the absence of scattering [9,10]. Thus, it was predicted that in addition to the impurity contribution to the spin-Hall conductivity, there must be a purely internal contribution, due only to the band structure of an ideal crystal.

So far, transport studies of SHE on single-crystal structures with macroscopic mean free paths of electrons, which minimize impurity mechanisms and, thus, correspond to the best conditions for the study of exactly intrinsic spin-dependent effects, have not been carried out. Filling this gap, we present the results of such a study, fundamentally different in its formulation from other transport studies SHE.

According to the relativistic momentum-spin dynamics of electrons, counter movement of the spin flows in the direction transverse with respect to the current direction accumulates spins of opposite polarization on opposite sides of the samples (see fig. 1). In the cross-section of the sample, symmetric with respect to the z-axis, the counter spin flows are equal and, without violating the equilibrium charge distribution along the y-axis, cannot create a transverse potential difference. For this reason, the study of SHE by electric methods is possible only for a nonequilibrium distribution of spins, and hence charges, as was achieved in known transport experiments [7] by

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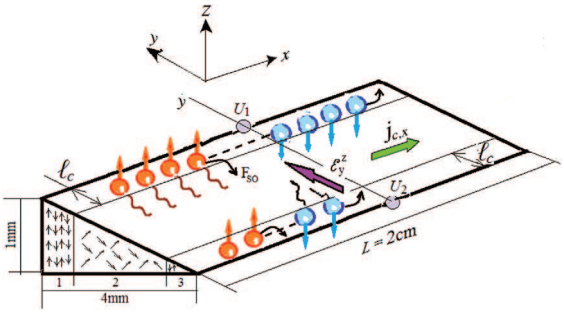

Fig. 1: (Colour online) Schematic representation of a sample with an asymmetric cross-section, leading to nonequilibrium dynamics and accumulation of spins in the presence of a spin-orbit interaction. Areas 1 and 3 refer to N_1 and N_3 numbers of carriers, respectively. The spin-flip region is symbolically marked by 2.

preliminary spin polarization of unpolarized injected current. Usually this was achieved with the help of ferromagnets, whose ability to create a spin polarization of the current is confirmed by observing in them an anomalous spin-Hall effect. The spin-polarized current obtained in the ferromagnet was then injected into the material under study, inducing charge and spin nonequilibrium (in particular, SHE) in this material, which was studied [11,12].

To avoid ambiguity in interpreting the final results, we created a nonequilibrium distribution of spins in the metal, without resorting to external methods of preliminary polarization of the current. Under the assumption of the existence of an SO interaction, this can be done with an asymmetric shape of the sample cross-section A with the characteristic size $\sqrt{A} \gg \ell_c$ (ℓ_c is the mean free path of carriers) on the width of the sample (fig. 1). A sample of aluminum wedge-shaped was cut by an electric-spark method from a high-purity ($\rho_{4,2\,\mathrm{K}} = 1,6 \cdot 10^{-10}\,\mathrm{Ohm \cdot cm}$) single-crystal ingot (made by zone melting at the Volkhov Aluminum Plant named after SM Kirov) and processed by chemical etching and polishing. The physics of the appearance of a nonequilibrium spin regime with the given geometry of the sample cross section in a semiclassical language can be described as follows.

In a perfect crystal, the carrier density n and the dc density $\mathbf{j}_{c,x} = ne\delta\mathbf{v}$ ($\delta\mathbf{v}$ is the drift velocity of carriers) are not related to the sample shape, while the total number of carriers depends on its volume. If the spin-orbit field removes the spin degeneracy, then, say, on the scale of the mean free path l_c ($\approx 0.1\,\mathrm{mm}$) from the edges, where the spins, in any case, preserve the orientation, the number of carriers on the left and on the right of the width of the sample of length L (areas 1 and 3 in fig. 1), in volumes $V_1 = A_1 L$ and $V_3 = A_3 L$ will be $N_1 = nV_1$ and $N_3 = nV_3$, respectively, with an equal number of spins of both orientations, "*up*" and "*down*", parallel to the spin-orbit field, so $N_1 \neq N_3$.

In the absence of a current $j_{c,x}$, the distribution of the spins and their corresponding charges is in equilibrium, and the gradients of the electrochemical potential and the spin chemical potential in the sample are absent in any directions. According to the SO interaction concept, the nonequilibrium dynamics of spins in momentum space should appear at $\delta\mathbf{v} \neq 0$ due to the induced additional addition to the spin-orbital field (for example, within the Hamiltonian with a Rashba-type spin-orbit coupling [8]), leading to a spin-current of two directions along y, depending on whether the spins are deflected upwards ($k_y > 0$) or downward ($k_y < 0$) (k is the quasimomentum). If the dimensions of the sample over the width significantly exceed the mean free path, as in our sample, then in the direction y there must exist a "spin-flip" region (shown in yellow in the figure), where orientations of the moving spins are is stochastized. As a result, the accumulation of spins at opposite edges of the sample, arising under the action of the spin current, turns out to be nonequilibrium, leading to the appearance of a transverse potential difference $U_y^s = U_{y1} - U_{y2}$, not related to the Lorentz force, and therefore independent of the direction commutation of the current $j_{c,x}$, and hence the direction of the gradient of the nonequilibrium *spin* chemical potential, $\nabla\mu_s$.

This allows us to distinguish and isolate U_y^s in the presence of the common usual Hall effect (CHE) in the case of a magnetic field, since, unlike the conserved U_y^s sign, the U_{CHE} sign, given by the direction of the Lorentz force, commutes to the opposite one as each of the directions both the current $j_{c,x}$ and H, in particular H_z, changes. The necessity of using a magnetic field, that, in general, intensifies SHE, is dictated by a very small value of the SHE as a second-order relativistic effect. A reasonable compromise between the value of CHE and the possibility of resolution, on its background, SHE, was achieved in small magnetic fields at $\omega_c\tau \ll 1$ (ω_c is the cyclotron frequency, τ is the relaxation time). In view of this, we worked in the interval of magnetic fields at $\omega_c\tau \leq 0.1$, which, as will be noted below, also has other important features.

Thus, using a magnetic field, on the Hall probes U_{y1} and U_{y2} located along the y-axis, we measured a potential difference consisting of two contributions —the CHE contribution, due to the Lorentz force, and the SHE contribution [13], caused by the nonequilibrium accumulation of spins under conditions of inhomogeneous distribution of spins due to the shape effect at macroscopic sample sizes:

$$U_H = U_{CHE} + U_{SHE} = \int_0^w \mathbf{E}_{CHE}\mathbf{j}_y \mathrm{d}y + \int_0^w \mathcal{E}_y^z \mathrm{d}y, \quad (1)$$

where

$$\mathbf{E}_{CHE} = (n^* e)^{-1}[\mathbf{B}_{\pm z} \times \mathbf{J}_{\pm x}] \quad (2)$$

and $\mathcal{E}_y^z$ is a Hall effective spin-dependent electric field [13, 14]

$$\mathcal{E}_y^z = \frac{\alpha}{\ell_c k_\mathrm{F}} [\hat{\sigma} \times \nabla\mu_c]_y. \quad (3)$$

In expressions (1)–(3), w is the size of the sample along the axis y; $\mathbf{B}$ and $\mathbf{J}$ are the induction vector and the current

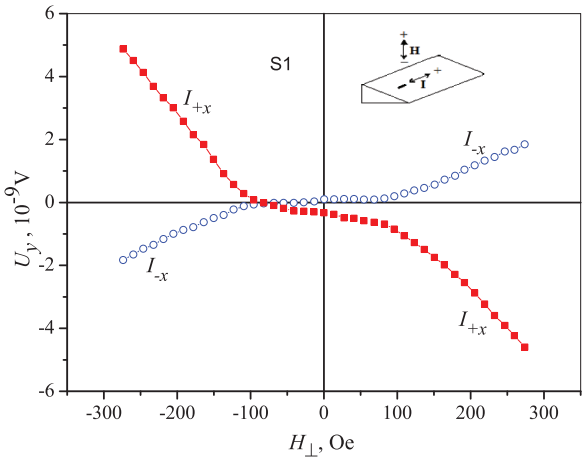

Fig. 2: (Colour online) The transverse potential difference U_y of sample S1 with a wedge-shaped cross-section as a function of the magnetic field H_z and $j_{c,x}$.

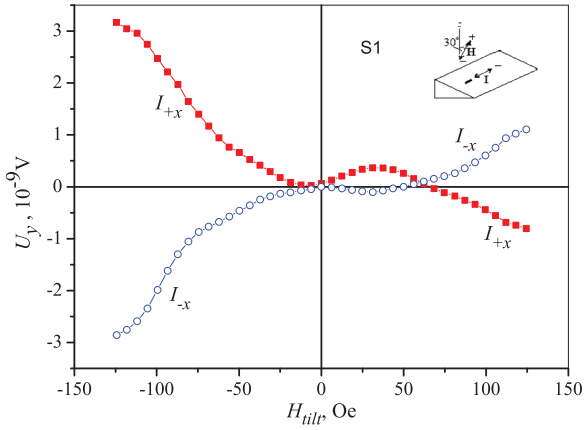

Fig. 3: (Colour online) The transverse potential difference U_y in the sample S1 as a function of the inclined magnetic field H_{tilt} and $j_{c,x}$.

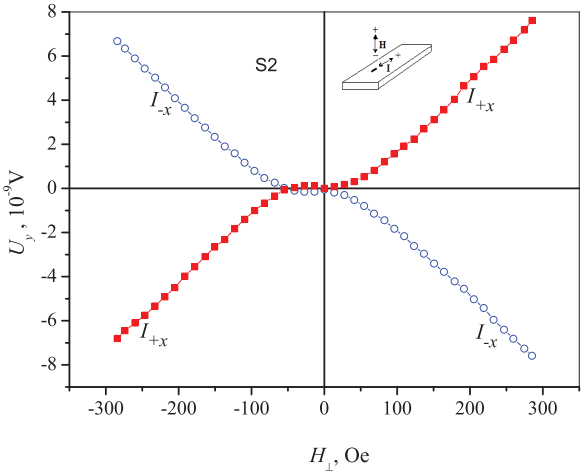

Fig. 4: (Colour online) The transverse potential difference U_y in sample S2 with a symmetric (rectangular) cross-section as a function of the magnetic field H_z and $j_{c,x}$.

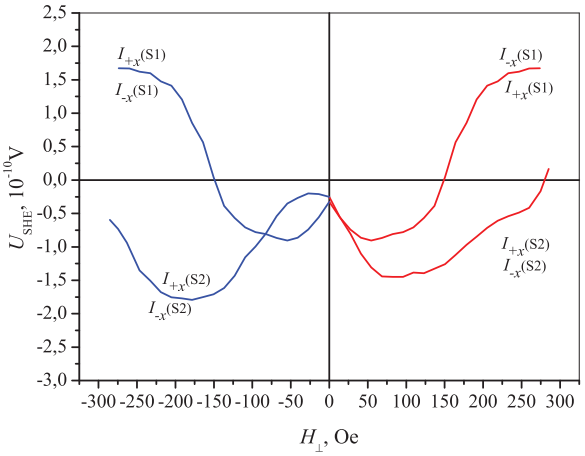

Fig. 5: (Colour online) Spin-Hall effect in aluminum samples with asymmetric (S1) and symmetrical (S2) cross-sections in a perpendicular magnetic field.

density vector, respectively; k_F is the wave Fermi vector; $\hat{\sigma}_y$ are the Pauli matrices; α is the spin-orbit coupling constant, μ_c is the chemical potential of the charge carriers, and n^* is the effective concentration of charge carriers; $\mathbf{j}_y$ is the unit vector of the y-axis.

Figures 2, 3, and 4 show the transverse potential difference U_y as a function of the magnetic field H for $\mathbf{J} \| \pm x$ and two orientations of $\mathbf{H}$ —parallel (figs. 2, 4) and nonparallel (fig. 3) to the axis z. The functions $U_{SHE}(H)$, found from the four equations, using the CHE sign reversal property for the commutation of directions of the vectors $\mathbf{H}$ and $\mathbf{J}$, are shown in fig. 5 for cases with asymmetric (S1) and symmetric (S2) configurations of the cross-section of the samples (see the insets to figs. 2, 4) with respect to the z-axis, and in fig. 6 for the case with a magnetic field inclined to that axis (see the inset to in fig. 3) for sample S1 (also, for comparison, SHE is shown in sample S2 in a perpendicular magnetic field).

Despite the difference between the curves in figs. 2–4 in detail, they nevertheless share a common property —the reversal of the sign when the direction of any of the vectors changes to the opposite. This means that the transverse charge imbalance in the samples studied due to the Lorentz force (the first term in eq. (1)) predominates and exceeds the charge imbalance given by the spin imbalance (the second term in eq. (1)) for sample S1 by several times (compare figs. 2 and 3 with figs. 5 and 6), and by at least two orders of magnitude for the sample S2 with a symmetrical cross-section (compare fig. 4 with the data for sample S2 in figs. 5 and 6).

Other behavior is demonstrated by the functions U_{SHE}, shown in figs. 5 and 6. It is clear that it has nothing to do with the behavior of U_{CHE}: the sign of these functions neither in the perpendicular nor in the inclined magnetic fields commutes either with a change in the direction of the current or with reversal of the direction (but not magnitude) of the magnetic field, which is in conceptual accordance with the above-described physics nature of the

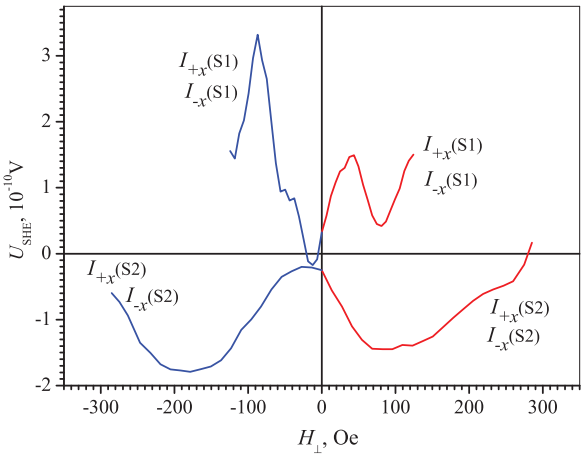

Fig. 6: (Colour online) Spin-Hall effect in an aluminum sample S1 in an inclined magnetic field. For comparison, SHE is shown in sample S2 in a perpendicular magnetic field.

spin-Hall effect. Moreover, the behavior of U_{SHE} surprisingly correlates with the well-known data on the band features of the dependence of the density of states (carrier concentration) on the magnetic field in Al.

It is well known that in trivalent aluminum, the effective carrier concentration in a magnetic field is determined by the mixed contribution from of Fermi carriers in the second (hole) and third (electron) energy bands [15]. Due to this, the effective carrier concentration in Al dramatically depends on the ratio of the contributions from carriers of different signs in the effective field range $0 < \omega_c\tau < 0.1$, changing the sign in this interval [15,16], or for our samples with $l_c \approx 0.1$ mm in the field interval 50–300 Oe. This fully explains the character of the nonlinearity observed in the curves U_{SHE} in figs. 5, 6, including the dependence of the sign U_{SHE} on the value of $\omega_c\tau$ in the above field interval. (We recall that the sign of the ordinary Hall voltage (as can be seen in figs. 2–4) does not change in this case when the sign of the carriers changes due to a change in the direction of the Lorentz force.) Thus, the nonlinear behavior of U_{SHE} in samples S1 and S2 indisputably testifies the manifestation of the band (intrinsic) spin-Hall effect in ultrapure aluminum. As for the experiment with an inclined magnetic field (see the inset to in fig. 6), the results of which are shown in fig. 6, then the main conclusion from this experiment is the absence of a noticeable difference between this case and that in the case with a perpendicular field.

Estimate of the spin-orbit coupling coefficient α for Al from the second term in eq. (1) according to the measured value of $U_{SHE} \simeq 2.5 \cdot 10^{-11}$ eV for $H = 0, k_F = 1.75 \cdot 10^8\,\mathrm{cm}^{-1}, l_c \approx 0.1$ mm, $\mu_c = 11.1$ eV gives the value $\alpha \approx 1.13 \cdot 10^{-5}$, which is two orders of magnitude smaller than the one given in the literature from the measurements of impurity effects [7,11].

In conclusion, the direct electrical measurements of the relativistic spin-Hall effect (SHE) due to the spin-orbit interaction were carried out for the first time. This is made possible by the artificially created, transverse to the transport current, spin-off imbalance induced by the shape effect. This imbalance, in its turn, induces the corresponding charge imbalance, which differs in its chiral features from the charge imbalance in the common Hall effect (CHE). For the first time, SHE measurements were carried out on a massive, ultra-pure nonmagnetic metal-aluminum single crystal. To identify the effect, we use the effective magnetic field range $0 < \omega\tau < 0.1$, where the carriers, and with them the effective concentration, change sign. The effect was studied at a liquid-helium temperature with a resolution of the potential difference $\sim 10^{-11}$ V [17]. The nonlinear behavior of the SHE detected in a magnetic field serves as a direct proof of the existence of the nonequilibrium dynamics of spins in momentum space and the manifestation of an intrinsic (band) mechanism for the spin polarization of electrons in a nonmagnetic metal under the conditions of a minimized external impurity spin polarization factor. The value of the spin-orbit coupling coefficient in aluminum obtained by us is two orders of magnitude smaller than that estimated from indirect measurements of SHE in disordered metallic nanostructures [10].

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