Dynamic nuclear spin polarization induced by Edelstein effect at Bi(111) surfaces

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Nuclear spin polarization induced by hyperfine interaction and the Edelstein effect due to strong spin-orbit interaction is investigated by quantum transport in Bi(111) thin film samples. The Bi(111) films are deposited on mica by van der Waals epitaxial growth. The Bi(111) films show micrometer-sized triangular islands with 0.39 nm step height, corresponding to the Bi(111) bilayer height. At low temperatures a high current density is applied for fixed durations to generate a non-equilibrium carrier spin polarization by the Edelstein effect at the Bi(111) surface, which then induces dynamic nuclear polarization by hyperfine interaction. Comparative quantum magnetotransport antilocalization measurements show that the quantum phase decoherence times decrease and the spin-orbit decoherence times increase as the polarization duration increases. Both outcomes indicate the suppression of antilocalization by the in-plane Overhauser field from the nuclear polarization and allow a quantification of the Overhauser field. Hence nuclear polarization was both achieved and quantified by a purely electronic transport-based approach.

Spin-orbit interaction (SOI) on surfaces and the corresponding splitting of surface-state bands have garnered wide attention [1]. Bi thin films possess an anisotropic Fermi surface characterized by large SOI, a low carrier density, high mobility, and a long mean free path [2]. Spatial inversion symmetry exists in the bulk but is broken normal to the surface yielding two Rashba-like surface spin subbands [3, 4]. The Bi (111) surface supports two-dimensional (2D) surface states which hence show strong Rashba-like SOI due to the asymmetry of the surface-confinement potential, with a Rashba parameter $\alpha_B \approx 0.5$ eV Å, an order of magnitude larger than in 2D electron systems in InAs and InSb heterostructures [5–7]. The Bi (111) surface states have therefore formed the subject of recent investigations to more fully understand their properties [8, 9].

The Edelstein (or Rashba-Edelstein) effect describes the generation of a non-equilibrium carrier spin polarization (CP) in materials with strong SOI in response to an applied electric field or a current density $j$, with the spin polarization direction normal to $j$ and the surface normal [10–13]. The Edelstein effect has its origin in spin-momentum locking, a characteristic feature of SOI. It has been shown that the Edelstein effect can be pronounced at surfaces and interfaces with strong SOI, such as the Ag/Bi(111) [14] and Cu/Bi(111) [15] interfaces. Given the strong SOI at the Bi(111) surface, an in-plane $j$ in a Bi thin film is expected to generate a non-equilibrium in-plane CP. In turn, hyperfine interaction (HI) can by dynamic nuclear polarization (DNP) transfer the CP to a non-equilibrium in-plane nuclear spin polarization (NP). The present work shows the existence of DNP via the Edelstein effect, an example of the interplay between strong SOI, HI, and the Edelstein effect. The work also demonstrates that the effect of NP on quantum-coherent transport allows for a quantification of the polarization. The work is further reminiscent of recent experiments where the CP originating in the Edelstein effect generates a spin torque on magnetic moments in magnetic layers [16], albeit presently with HI replacing the spin torque action.

HI refers to the coupling of carrier spins to the nuclear spins by an energy term $A J \cdot I$, where $A$ represents the hyperfine coupling constant [17, 18], $I$ the nuclear spin and $J$ the total carrier angular momentum. Two mechanisms contribute to HI [18, 20], Fermi contact interaction (dominant when carrier and nuclear orbitals overlap [21]) and dipolar interaction (favoring carrier orbitals with a node at the nucleus [18, 19]). HI can be more pronounced for heavy atoms featuring atomic parameters with higher energy scales [17, 20], and for nuclei with large $I$. Both effects play a role strengthening HI for Bi, with $I = 9/2$. Further, electrons in Bi have a substantial s-orbital component at the Fermi energy, $\sim 10\%$, increasing the contact term and HI due to non-zero carrier density at the nucleus. The strong SOI in Bi may also enhance HI. Quantitative information on the strength of HI in semimetallic Bi is lacking, partially remediable by the approach presented here. Yet experiments have studied the interaction between Bi donors in Si and the Si s-like conduction band carriers [20, 22, 23], concluding $A = 6.1$ μeV. The Knight shift in Bi$_2$Se$_3$ indicates $A = 27$ μeV [17]. Such values for $A$ indicate that consequential HI is expected in semimetallic Bi as well as in Bi compounds. As mentioned, HI can lead to DNP where spin polarization is transferred from the carriers to the nuclei [24, 25] and CP then generates NP. With NP established, the carriers experience HI as an effective in-plane magnetic field having the same effect as an external Zeeman field [18, 25, 20], the Overhauser field $B_{OH}$.

$B_{OH}$ and the NP are in this work quantified by the weak-localization quantum coherence corrections to the conductance of the Bi(111) surface states, caused by quantum interference between backscattered time-reversed carrier trajectories. At low temperatures $T$, the quantum coherence corrections lead to a resistance $R$ with a specific dependence on an external magnetic field $B_\perp$ applied normally to the surface, in the absence of SOI known as weak-localization (WL) and under strong SOI known as (weak-)antilocalization (AL) [27, 30]. The magnetoconductance (MR, $R(B_\perp)$) due to AL is here determined by three characteristic times [27, 28, 30]: the elastic scattering time $\tau_0$ as deduced from the areal surface state density $N_S$ and mobility $\mu$, the SOI spin decoherence time $\tau_{SO}$, and the quantum phase decoherence time $\tau_\phi$. Here $\tau_{SO} \propto \Delta^2_{SO}$ where $\Delta_{SO}$ denotes the SOI splitting at the Fermi wavevector. The times are experimentally deter-
mensioned by quantitative fitting of the MR data to the AL theory developed by Iordanskii, Lyanda-Geller and Pikus (ILP) [31] appropriate for the Bi(111) 2D surface states with Rashba-like SOI. The influence of magnetization on AL in ferromagnetic materials has been theoretically studied [32]. We expect similar effects due to NP, supported by the theoretical treatment of $B_{OH}$ as an effective in-plane magnetic field [33] [34]. In particular, $B_{||}$ generates an effective Zeeman splitting which tends to align the carrier spins and hence suppresses the Cooperon in the spin singlet channel and thereby inhibits AL in favor of WL [32]. The inhibition of AL is visible in the data as an increase in $\tau_{SO}$ with increasing $B_{||}$. Further, AL is a sensitive probe of quantum and spin coherence [27] [29], and is sensitive to the time-reversal symmetry (TRS) breaking due to $B_{||}$ [32] [35] [36]. The breaking of TRS due to the interplay of Zeeman splitting and SOI results in a quantifiable decrease in $\tau_{0}$ [35] with increasing $B_{||}$, also visible in the data. Identifying $B_{||} = B_{OH}$, we thus use AL as a sensitive probe of DNP and HI. The values of $B_{OH}$ obtained by DNP via the Edelstein effect are calculated from a quantitative analysis of AL.

An optimized van der Waals epitaxy (vdWE) [37] was used to grow the Bi(111) films on mica substrates, resulting in large grain sizes with the trigonal axis perpendicular to the film plane, as described in Supplemental Material [38]. vdWE is particularly suited to the unstrained growth of weakly bonding materials such as Bi [39] [40]. The 40 nm thick Bi(111) was deposited through a shadowmask, yielding samples of diameter $\sim 350 \mu m$. Au contacts were photolithographically patterned after film deposition (Fig. 1b). Atomic force microscopy clearly indicated a layered step surface with triangular terraces (Fig. 1d) and showed a step height between adjacent terraces of $0.391 \pm 0.015 \mu m$, corresponding to one Bi(111) bilayer height [BL$_{111} = 0.39 \mu m$] [43].

The AL and transport coefficient characterization were carried out by magnetotransport in a $^3$He cryostat down to $T = 0.39 K$, using standard 4-contact AC lock-in techniques with current of 2 $\mu A$ rms under applied $B_{L}$. To develop DNP a high DC polarization current, $I_p = 0.5 mA$ to 2 mA, $j \sim 6.25 \times 10^7 \text{ A/m}^2$ to $2.5 \times 10^8 \text{ A/m}^2$, was applied at $T = 0.39 K$ between a pair of contacts for variable polarization durations $t_p$ from 10 to 120 min. $I_p$ was removed after the DNP step, letting the NP and $B_{OH}$ decay slowly with a spin-lattice relaxation time $T_1$ characteristic of the nuclear decoherence [41] [42]. The slow decay allowed time for the subsequent observation of DNP from AL measurements. For AL measurements the voltage was measured over the same contacts used for AL measurements the voltage was measured over the same contacts used for AL. $N_S$ and $\mu$ were determined from magnetotransport at 0.39 K, and the results indicate predominantly n-type surface carrier contribution. We determine $N_S = 1.95 \times 10^{15} \text{ m}^{-2}, \mu = 1.00 \text{ m}^2/\text{Vs}$ and correspondingly $\tau_0 = 856 \text{ ps}$ and mean free path $l_0 = v_f \tau_0 = 20.4 \text{ nm}$, where $v_f$ is the Fermi velocity derived from $N_S$. As appropriate for conductance dominated by surface states we use the 2D diffusion constant $D$ calculated as $D = \frac{1}{2}v_f^2\tau_0$ at $T = 0.39 K$ yielding $D = 0.00243 \text{ m}^2/\text{s}$. AL results in a characteristic positive quantum correction in $R(B_{L})$ at $B_{L} \lesssim 0.4 \text{ T}$, expressed as a small correction to the 2D conductivity $\sigma_2(B)$. We define $\Delta\sigma_2(B_{L}) = \sigma_2(B_{L}) - \sigma_2(B_{L} = 0)$ and $\Delta R(B_{L}) = R(B_{L}) - R_0$ where $R_0 = R(B_{L} = 0)$. Since $\Delta R(B_{L}) \ll R_0$, we have $\Delta\sigma_2(B_{L})/\sigma_2(B_{L} = 0) \approx -\Delta R(B_{L})/R_0$, allowing fits to $\Delta\sigma_2(B_{L})$ from the experimental MR. To fit the data ILP theory [31] is applied, including only the Rashba SOI term. Since $\tau_0$ merely produces a shift in $\Delta\sigma_2(B_{L})$, $\tau_0$ and $\tau_{SO}$ are the only two free fitting parameters. The fits are performed for AL obtained after different $t_p$ under $I_p = 1 \text{ mA}$. From the fits, we find the dependence on $t_p$ of $\tau_{SO}$ and $\tau_0$. From the latter the dependence on $t_p$ of $B_{OH}$ is determined.

Figure 3 depicts the MR of the Bi film sample at $T = 0.39 K$ before and after DNP using variable $I_p$ ranging from 0.5 mA to 2.0 mA and $t_p$ ranging from 0 (before DNP) to 120 min. The positive MR characteristic of AL is observed both before and after DNP. The negative of $\Delta\sigma_2(B_{L})$ (reproducing $\Delta R(B_{L})$) at low $B_{L}$ is displayed in Fig. 4 for variable...
$t_p$ when $I_p = 1$ mA. Best fits to the ILP theory \[31\] overlay the data in Fig. 4b in red and indicate that the theory excellently captures the AL in Bi(111) surface states with Rashba-like SOI and will allow reliable extraction of values for $\tau_{SO}$ and $\tau_\phi$. The traces for $R(B_\perp)$ (Fig. 5) and for $\Delta \sigma_2(B_\perp)$ (Fig. 4b) show a widening vs $B_\perp$ for $B_\perp \neq 0$ after DNP, characteristic of an increase in $\tau_{SO}$ (decreasing effect of SOI) and a decrease in $\tau_\phi$ as quantitative analysis will confirm. The widening shows a dependence on $I_p$ and $t_p$, with a long $t_p = 120$ min at $I_p = 1$ mA resulting in the largest effect. The dependence on $t_p$ and $I_p$ suggests DNP and hence $B_{OH}$ play a role in changing $\tau_{SO}$ and $\tau_\phi$. The widening of the minimum in $\Delta \sigma_2(B_\perp)$ is further illustrated in Fig. 5b where the black trace represents $\Delta \sigma_2(B_\perp)$ before DNP and the blue trace after DNP with $t_p = 60$ min and $I_p = 1$ mA. Before we present quantitative data on $\tau_{SO}$ and $\tau_\phi$, we note that the AL results after DNP are qualitatively consistent with the existence of in-plane $B_{OH}$. Phenomenologically, after removing $I_p$, $B_{OH}$ persists and generates an effective Zeeman energy $g_\parallel \mu_B B_{OH}$, where $g_\parallel$ denotes the in-plane $g$-factor (for Bi(111) surface states, $g_\parallel \approx 33$ [29]) and $\mu_B$ denotes the Bohr magneton. $B_{OH}$ partially aligns the carrier spins and suppresses the spin phase shift due to SOI and thereby weakens AL [32,35,45]. The effect leads to a widening of the characteristic sharp minimum in $\Delta R(B_\perp)$ vs $B_\perp$ and is quantified by a lengthening of $\tau_{SO}$. Further, $B_{OH}$ results in a spin-induced TRS breaking [34,35,45], leading to a decrease in $\tau_\phi$. Future theoretical studies specific to the influence of HI and NP on AL may help refine quantitative aspects of the experiments, as was performed for ferromagnetic order [32] and for Zeeman interaction [34].

The dependences of $\tau_{SO}$ and $\tau_\phi$ on $t_p$ at fixed $I_p = 1$ mA are presented in Fig. 5. The value of $\tau_{SO}$ increases with increasing $t_p$ (Fig. 5a), indicative of the influence of the in-plane $B_{OH}$ generated by DNP of the Bi nuclei. A phenomenological understanding was presented above. Theoretical studies of the combined influence of SOI and $B_{||}$ on the evolution of an inhomogeneous spin distribution at an interface [40] show that even a weak $B_{||}$ results in a decrease of the spin density proportional to $1/(2\pi D \tau_{SO})$, relating an increase in $\tau_{SO}$ to the influence of $B_{||} = B_{OH}$. We note that in Fig. 5 the data-point at $t_p = 60$ min was obtained after a different cooldown, and hence under different sample conditions than the other datapoints, explaining its outlying character (indicated by its different color). The maximum observed for $\tau_{SO}$ vs $t_p$ and the corresponding minimum for $\tau_\phi$ are thus regarded as artefacts within the accuracy of the data, and rather a saturation at higher $t_p$ should be inferred. Figure 5b shows a decrease of $\tau_\phi$ with increasing $t_p$, indicative of the interplay of the effective Zeeman energy and SOI [34,35]. The interplay is predicted to result in a quadratic dependence of $\tau_\phi$ on $B_{||}$ [35].
\[ \frac{\tau_0(B)}{\tau_0(B|| = 0)} = \frac{1}{1 + c B^2} \]

where \( c \) is expressed as

\[ c = \tau_0(B|| = 0) \tau_{SO}(B|| = 0)(g^*_{\mu_B}/\hbar)^2 \]

The estimated average value of \( B_{OH} = B|| \) can be calculated from the data using Eqs. (1-2), and plotted vs \( t_p \) after DNP under fixed \( I_p = 1 \) mA. Since the AL measurement (sweeping over \( B_\perp \sim 0.2 \) T after removing \( I_p \)) has a duration of \( \sim 15 \) min, by estimated average \( B_{OH} \) is meant the value calculated averaging over \( \sim 15 \) min decay. Current spreading between the two current contacts over the sample geometry during DNP will likely lead to non-uniform DNP, and \( B_{OH} \) hence encompasses spatial averaging as well. As plotted in Fig. 6 this average \( B_{OH} \) increases with increasing \( t_p \), and saturates at about 13 mT after \( t_p \approx 60 \) min (the datapoint at \( t_p = 60 \) min again in a different color as outlined above). The steady increase towards saturation of \( B_{OH} \) vs \( t_p \) and the saturation after tens of min are both consistent with expectations for DNP in Bi. The maximal \( B_{OH} \) obtainable, assuming full NP and full wavefunction overlap, can be estimated as \( B_{OH,max} = A/(g^*_{\mu_B}) \) \[\text{(17)}\] using values of \( A = 6.1 \) \( \mu \text{eV} \) to 27 \( \mu \text{eV} \) \[\text{(17)}\text{,}\text{ (20)}\text{,}\text{ (22)}\text{,}\text{ (23)}\] we find \( B_{OH,max} \approx 14.5 \) mT to 64 mT. Since we do not expect full NP and full wavefunction overlap, and since the obtained \( B_{OH} \) values involve estimates described above, the saturation value of 13 mT is consistent with the present knowledge of \( A \) in Bi. Hence, both the dependence of \( t_p \) and the saturation value strongly suggest that the CP due to the Edelstein effect in the Bi(111) surface states was transferred by HI to the Bi nuclei, demonstrating Edelstein-induced DNP. The measurements also illustrate that quantum transport in the form of AL forms a sensitive probe of NP. Beyond illustrating the interplay between strong SOI, HI, and the Edelstein effect, the Edelstein-induced DNP may find application as method to polarize nuclei to mitigate the effect of spin decoherence via HI in quantum devices \[\text{(24)}\text{,}\text{ (43)}\].

In conclusion, Bi(111) thin films deposited by van der Waals epitaxy on mica substrates show 0.39 nm step heights corresponding to one Bi(111) bilayer. Using antilocalization quantum-coherent transport measurements on the Bi(111) surface states to probe the existence of in-plane magnetic fields, quantitative experimental evidence was obtained for a transfer of carrier spin polarization to Bi nuclear spin polarization by hyperfine interaction. The carrier spin polarization was obtained via the Edelstein effect in the Bi(111) surface states with strong spin-orbit interaction. The experiments verify the existence of Edelstein-induced dynamic nuclear polarization, in an example of interaction between spin-orbit interaction and hyperfine interaction via the nuclear spin bath. The experiments also show that antilocalization measurements form a sensitive probe for hyperfine interaction and nuclear polarization. The work was supported by the U.S. Department of Energy, Office of Basic Energy Sciences, Division of Materials Sciences and Engineering under award DOE DE-FG02-08ER46532.

\[ \text{FIG. 6: Overhauser field } B_{OH} \text{ at } T = 0.39 \text{ K obtained at fixed DNP current } I_p = 1 \text{ mA, plotted vs DNP duration } t_p. \text{ The line is a guide to the eye.} \]

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