## Constraints on Exotic Spin-Dependent Interactions Between Matter and Antimatter from Antiprotonic Helium Spectroscopy

Filip Ficek, <sup>1,\*</sup> Pavel Fadeev, <sup>2,3</sup> Victor V. Flambaum, <sup>2,4</sup> Derek F. Jackson Kimball, <sup>5</sup>
Mikhail G. Kozlov, <sup>6,7</sup> Yevgeny V. Stadnik, <sup>2</sup> and Dmitry Budker <sup>2,8,9</sup>

<sup>1</sup>Institute of Physics, Jagiellonian University, Łojasiewicza 11, 30-348 Kraków, Poland <sup>2</sup>Helmholtz Institute Mainz, Johannes Gutenberg University, 55099 Mainz, Germany <sup>3</sup>Ludwig-Maximilians-Universität, München, Fakultät für Physik, Arnold Sommerfeld Center for Theoretical Physics, 80333 München, Germany

<sup>4</sup>School of Physics, University of New South Wales, Sydney, New South Wales 2052, Australia <sup>5</sup>Department of Physics, California State University—East Bay, Hayward, California 94542-3084, USA <sup>6</sup>Petersburg Nuclear Physics Institute of NRC "Kurchatov Institute", Gatchina 188300, Russia <sup>7</sup>St. Petersburg Electrotechnical University LETI, Prof. Popov Str. 5, 197376 St. Petersburg, Russia <sup>8</sup>Department of Physics, University of California at Berkeley, Berkeley, California 94720-7300, USA <sup>9</sup>Nuclear Science Division, Lawrence Berkeley National Laboratory, Berkeley, California 94720, USA

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Heretofore undiscovered spin-0 or spin-1 bosons can mediate exotic spin-dependent interactions between standard model particles. Here, we carry out the first search for semileptonic spin-dependent interactions between matter and antimatter. We compare theoretical calculations and spectroscopic measurements of the hyperfine structure of antiprotonic helium to constrain exotic spin- and velocity-dependent interactions between electrons and antiprotons.

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Antiprotonic helium (He $^+\bar{p}$ ) is a helium atom where one of the electrons is replaced with an antiproton. Antiprotonic helium, being a relatively simple matter-antimatter bound state, can provide insight into possible exotic matterantimatter interactions. Since the first observations of relatively long-lived (lifetimes of the order of microseconds) antiprotonic helium atoms in 1991 [1], there have been significant developments in experimental techniques. The latest achievements include determining the antiproton magnetic moment [2] (this result, utilizing the antiprotonic helium, was improved later with direct measurements [3,4]), resolving the hyperfine structure of  ${}^{3}\text{He}^{+}\bar{p}$  [5] and precise measurements of the antiproton-to-electron mass ratio [6]. Furthermore, theoretical calculations of transition energies in antiprotonic helium have reached agreement with the experiment at a level of one part in  $10^9$  or better in many cases [7–11]. An extensive summary of research on antiprotonic helium prior to 2002 can be found in Ref. [12].

The principal focus of antimatter research to date has been on tests of *CPT* invariance [13], for example, by measuring the properties of the antiproton [2–4,6], and on constraining Yukawa-type spin-independent forces [14,15]. In this Letter, we show that one can also search for exotic spin-dependent interactions between matter and antimatter from precise measurements and QED-based calculations of antiprotonic helium.

Spin-dependent interactions [16,17] (see also Ref. [18] for a review) appear in theories including "new," i.e., so far

undiscovered bosons such as axions [19–24], familons [25,26], Majorons [27,28], arions [29], new spin-0 or spin-1 gravitons [30–33], Kaluza-Klein zero modes in string theory [34], paraphotons [35–37], and new Z' bosons [38–40]. These new bosons are introduced to solve problems such as the nature of dark matter [41] and dark energy [42,43], the strong-CP problem [16], and the hierarchy problem [44].

The most commonly employed framework for the purpose of comparing different experimental searches for exotic spin-dependent interactions is that introduced in Ref. [16] to describe long-range spin-dependent potentials associated with the axion and later extended in Ref. [17] to encompass long-range potentials associated with any generic spin-0 or spin-1 boson. Some issues related to the velocity-dependent potentials presented in Ref. [17] were pointed out in Ref. [45]. The spin-dependent potentials enumerated in Ref. [17] are characterized by dimensionless coupling constants that specify the strength of the interaction between various particles and a characteristic range parameter \( \) for the interaction associated with the reduced Compton wavelength of the new boson of mass  $m_0$ ,  $\lambda = \hbar/(m_0 c)$ , where  $\hbar$  is the reduced Planck's constant and c is the speed of light. Depending on the nature of the new interaction, different particles will generally have different coupling constants.

To date, the constraints on exotic spin-dependent interactions between matter and antimatter have concerned

leptonic interactions, and they are derived from hydrogen-like atoms: positronium [46–48] and muonium [48–50]. In the following, we constrain spin-dependent interactions between an electron and an antiproton (a semileptonic interaction). We do this in a similar manner to Ref. [45], by comparing experimental results for the hyperfine structure of  ${}^4\text{He}^+\bar{p}$  [2] and QED-based calculations [51] and using our calculated expectation values of spin-dependent potentials.

The structure of this Letter is as follows. We begin by constructing approximate wave functions describing the antiprotonic helium atom. Then we present the relevant exotic potentials. Finally, we use first-order perturbation theory on the aforementioned wave functions and potentials to obtain constraints on the interaction parameters of interest.

Since the electron mass  $m_e$  is much smaller than the nuclear (alpha particle) and antiproton masses,  $m_{\rm nucl}$  and  $m_{\bar p}$ , respectively, the approximate Hamiltonian describing antiprotonic helium has the form [52]:

$$\hat{H} = \left(-\frac{\hbar^2}{2\mu_{\bar{p}}}\nabla_{\bar{p}}^2 - \frac{2e^2}{|\mathbf{r}_{\bar{p}}|}\right) + \left(-\frac{\hbar^2}{2m_e}\nabla_e^2 - \frac{2e^2}{|\mathbf{r}_e|}\right) + \frac{e^2}{|\mathbf{r}_{\bar{p}} - \mathbf{r}_e|},\tag{1}$$

where e is the elementary charge,  $\mu_{\bar{p}} = m_{\rm nucl} m_{\bar{p}} / (m_{\rm nucl} + m_{\bar{p}})$  is the reduced mass of the antiproton,  ${\bf r}_{\bar{p}}$  and  ${\bf r}_e$  are the position vectors of the electron and antiproton with respect to the nucleus (Fig. 1), and  $\nabla_{\bar{p}}$  and  $\nabla_e$  are Laplacians in the coordinates  ${\bf r}_{\bar{p}}$  and  ${\bf r}_e$ .

The strength of any hypothetical exotic spin-dependent interaction between two charged particles is orders-of-magnitude smaller than their electromagnetic interaction. Based on this, a high-precision calculation of the perturbation due to exotic effects is not required, and it is sufficient to calculate the exotic contributions to first order in perturbation theory. For these calculations, a relatively simple form of the approximate wave functions of the antiproton and electron may be assumed. In the following, we focus on antiprotonic helium with the antiproton in the (n, l) = (37, 35) state and the electron in the (1,0) state (where the first number in an ordered pair is the principal quantum number, and the second one is the orbital angular

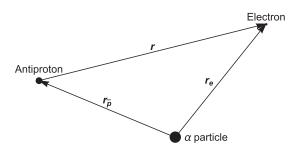


FIG. 1. Schematic diagram of the antiprotonic helium atom. The nucleus is an alpha particle.

momentum), since there are both relatively precise experimental data and theoretical calculations available for this system [2,51]. We use the approximate spatial wave function [53]

$$\Psi_{\tilde{m}_{\bar{p}}}(\mathbf{r}_{\bar{p}}, \mathbf{r}_{e}) = \frac{1}{\sqrt{1 - \beta^{2}}} \left[ \psi_{37,35,\tilde{m}_{\bar{p}}}^{(\bar{p})}(\mathbf{r}_{\bar{p}}) \psi_{1,0,0}^{(e)}(\mathbf{r}_{e}) - \beta \psi_{36,35,\tilde{m}_{\bar{p}}}^{(\bar{p})}(\mathbf{r}_{\bar{p}}) \psi_{1,0,0}^{(e)}(\mathbf{r}_{e}) \right], \tag{2}$$

where  $\beta$  is a numerical constant and  $\psi_{n,l,\tilde{m}}^{(a)}$  is a generalized hydrogenlike atom wave function [54] for a particle a with a principal quantum number n, an orbital angular quantum number l, and a magnetic quantum number  $\tilde{m}$ :

$$\psi_{n,l,\tilde{m}}^{(a)}(r,\theta,\phi) = \sqrt{\frac{4(Z_n^{(a)})^3 \mu^3 (n-l-1)!}{n^4 (n+l)!}} \left(\frac{2Z_n^{(a)} \mu^{(a)} r}{n}\right)^l \times e^{-\frac{Z_n^{(a)} \mu^{(a)} r}{n}} L_{n-l-1}^{2l+1} \left(\frac{2Z_n^{(a)} \mu^{(a)} r}{n}\right) Y_l^{\tilde{m}}(\theta,\phi).$$
(3)

In formula (3),  $\mu^{(a)}$  denotes the reduced mass of particle a,  $(\mu^{(\bar{p})} = \mu_{\bar{p}})$ ,  $\mu^{(e)} \approx m_e$ ,  $Z^{(a)}$  is the effective charge seen by particle a in a state with a principal quantum number n,  $L_{n-l-1}^{2l+1}$  is the generalized Laguerre polynomial, and  $Y_l^m$  is the spherical harmonic function. The parameters  $\beta$  and  $Z_n^{(a)}$  are derived using the variational method [57].

To get a full approximate wave function of the considered system, we need to add the spinor component to the spatial wave function (2). In the (37,35) state, the total orbital angular momentum of the atom is  $\mathcal{L} = 35$ . Let us then denote by  $|35, \tilde{m}_{\mathcal{L}}\rangle$  the vector corresponding to the spatial wave function  $\Psi_{\tilde{m}_{\tilde{p}}}.$  The interaction between the orbital motion and the electron spin is the strongest among the angular-momentum-dependent interactions in antiprotonic helium [7], so we first add the orbital angular momentum to the electron spin, obtaining  $\mathcal{F} = \mathcal{L} + s_e$ . We then include the antiproton's spin to obtain the total angular momentum  $\mathcal{J} = \mathcal{F} + s_{\bar{p}}$  [58]. This addition scheme introduces the hyperfine structure shown in Fig. 2. We may characterize any hyperfine state in the (37,35) manifold using three numbers:  $\mathcal{J}$ ,  $\tilde{m}_{\mathcal{J}}$ , and  $\mathcal{F}$ . We build these states using the Clebsh-Gordan coefficients  $C^{J,m_J}_{j_1,m_1;j_2,m_2}$ :

$$\begin{split} |\mathcal{J}, \tilde{m}_{\mathcal{J}}; \mathcal{F}\rangle &= \sum_{\tilde{m}_{\mathcal{F}}, \tilde{m}_{s_{\bar{p}}}} C_{\mathcal{F}, \tilde{m}_{\mathcal{F}}; s_{\bar{p}}, \tilde{m}_{s_{\bar{p}}}}^{\mathcal{J}, \tilde{m}_{\mathcal{J}}} |\mathcal{F}, \tilde{m}_{\mathcal{F}}\rangle |s_{\bar{p}}, \tilde{m}_{s_{\bar{p}}}\rangle \\ &= \sum_{\tilde{m}_{\mathcal{F}}, \tilde{m}_{s_{\bar{p}}}} \sum_{\tilde{m}_{\mathcal{L}}, \tilde{m}_{s_{e}}} C_{\mathcal{F}, \tilde{m}_{\mathcal{F}}; s_{\bar{p}}, \tilde{m}_{s_{\bar{p}}}}^{\mathcal{F}, \tilde{m}_{\mathcal{F}}} C_{L, \tilde{m}_{\mathcal{L}}; s_{e}, \tilde{m}_{s_{e}}}^{\mathcal{F}, \tilde{m}_{\mathcal{F}}} \\ &\times |35, \tilde{m}_{\mathcal{L}}\rangle |s_{e}, \tilde{m}_{s_{e}}\rangle |s_{\bar{p}}, \tilde{m}_{s_{\bar{p}}}\rangle. \end{split} \tag{4}$$

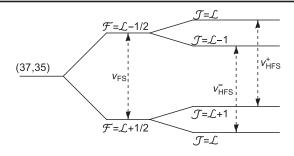


FIG. 2. Hyperfine structure of the (n, l) = (37, 35) state of an antiprotonic helium atom. The transitions denoted by  $\nu_{HFS}^{\pm}$  were investigated in Ref. [2].

Because of the rotational invariance of the Hamiltonian and the exotic spin-dependent potentials considered below, the respective matrix elements do not depend on the specific  $\tilde{m}_{\mathcal{J}}$  value, so we denote the hyperfine structure states by  $|\mathcal{J}; \mathcal{F}\rangle$ .

Reference [2] presented the results of measurements of energies for the transitions  $|35;35.5\rangle \leftrightarrow |34;34.5\rangle$  (denoted as  $\nu_{HFS}^-$ ) and  $|36;35.5\rangle \leftrightarrow |35;34.5\rangle$  (denoted as  $\nu_{HFS}^+$ ) along with the theoretically predicted values calculated in Refs. [51]. We compare them in Table I and present the values of  $\Delta E$ —a quantity constraining exotic interactions at a 90% acceptance level. We define it in such a way that

$$\int_{-\Delta E}^{+\Delta E} \frac{1}{\sqrt{2\pi}\sigma} e^{-(x-\mu)^2/(2\sigma^2)} dx = 0.9,$$
 (5)

where  $\mu$  is the mean difference between theoretical and experimental transition energies and  $\sigma^2 = \sigma_{th}^2 + \sigma_{exp}^2$  ( $\sigma_{th}$  and  $\sigma_{exp}$  are here theoretical and experimental uncertainties, respectively). These values of  $\Delta E$ , characterizing the level of agreement between theory and experiment, taking into account the uncertainties of both, are used to constrain the exotic interactions.

In Ref. [17], Dobrescu and Mocioiu introduced 16 independent spin-spin interactions. For studies of exotic spin couplings using  ${}^{4}\text{He}^{+}\bar{p}$ , only those interactions that are invariant under spatial inversion and time reversal are relevant. These two conditions allow shifts of energy levels in first-order perturbation theory. There are five spin-dependent potentials that satisfy these requirements: two

velocity-independent potentials and three velocity-dependent potentials. In the coordinate-space representation they have the form [45]

$$V_2 = f_2^{e\bar{p}} \frac{\hbar c}{\pi} (\mathbf{s}_{\bar{p}} \cdot \mathbf{s}_e) \frac{e^{-r/\hbar}}{r}, \tag{6}$$

$$V_{3} = f_{3}^{e\bar{p}} \frac{\hbar^{3}}{\pi m_{e}^{2} c} \left[ \mathbf{s}_{\bar{p}} \cdot \mathbf{s}_{e} \left( \frac{1}{\lambda r^{2}} + \frac{1}{r^{3}} + \frac{4\pi}{3} \delta^{3}(r) \right) - (\mathbf{s}_{\bar{p}} \cdot \mathbf{r}) (\mathbf{s}_{e} \cdot \mathbf{r}) \left( \frac{1}{\lambda^{2} r^{3}} + \frac{3}{\lambda r^{4}} + \frac{3}{r^{5}} \right) \right] e^{-r/\lambda}, \quad (7)$$

$$V_{4-5} = f_{4-5}^{e\bar{p}} \frac{i\hbar^3}{4m_e^2 c} \mathbf{s}_{\bar{p}} \cdot \left[ \left( \frac{m_e}{m_{\bar{p}} + m_e} \nabla_{\bar{p}} - \frac{m_{\bar{p}}}{m_{\bar{p}} + m_e} \nabla_e \right) \times \mathbf{r}, \right.$$

$$\left. \left( \frac{1}{r^3} + \frac{1}{\chi r^2} \right) e^{-r/\chi} \right]_+, \tag{8}$$

$$V_{4+5} = f_{4+5}^{e\bar{p}} \frac{i\hbar^3}{4m_e^2 c} \mathbf{s}_e \cdot \left[ \left( \frac{m_e}{m_{\bar{p}} + m_e} \nabla_{\bar{p}} - \frac{m_{\bar{p}}}{m_{\bar{p}} + m_e} \nabla_e \right) \times \mathbf{r}, \right. \\ \left. \left( \frac{1}{r^3} + \frac{1}{\chi r^2} \right) e^{-r/} \right]_+,$$
 (9)

$$V_{8} = -f_{8}^{e\bar{p}} \frac{\hbar^{3}}{4\pi m_{e}^{2}c} \left[ \mathbf{s}_{e} \cdot \left( \frac{m_{e}}{m_{\bar{p}} + m_{e}} \nabla_{\bar{p}} - \frac{m_{\bar{p}}}{m_{\bar{p}} + m_{e}} \nabla_{e} \right), \right.$$

$$\left[ \mathbf{s}_{\bar{p}} \cdot \left( \frac{m_{e}}{m_{\bar{p}} + m_{e}} \nabla_{\bar{p}} - \frac{m_{\bar{p}}}{m_{\bar{p}} + m_{e}} \nabla_{e} \right), \frac{e^{-r/\bar{\lambda}}}{r} \right]_{+} \right]_{+},$$

$$(10)$$

where  $f_i^{e\bar{p}}$  is the dimensionless coupling parameter of the ith interaction between the electron and the antiproton,  $\mathbf{r} = \mathbf{r}_e - \mathbf{r}_{\bar{p}}$  is the position vector directed from the antiproton to the electron, r is the distance between the electron and antiproton,  $\nabla_{\bar{p}}$  and  $\nabla_e$  are the vector differential operators in the coordinate space of the antiproton and the electron, respectively, and  $\mathbf{s}_{\bar{p}}$  and  $\mathbf{s}_e$  are the spins of the antiproton and the electron, respectively. By  $[\cdot,\cdot]_+$  we denote an anticommutator. Let us point out that the mass ratios present in the velocity-dependent potentials, appear in their derivation [45] when one considers particles with different masses. We also study  $V_{4\pm5}$  rather than

TABLE I. Experimental and theoretical transition energies between hyperfine-structure states in the (n, l) = (37, 35) manifold, along with their differences and values of  $\Delta E$ , a parameter describing the level of agreement between theoretical and experimental results and taking into account their uncertainties. We define  $\Delta E$  at the 90% Confidence Level (C.L.) in Eq. (5).

	Experiment [2]	Theory [51]	Difference	ΔE (at 90% C.L.)
$\overline{ u^+_{ m HFS}}$	12.896 641(63) GHz	12.8963(13) GHz	0.3(1.3) MHz	2.2 MHz
$ u_{\mathrm{HFS}}^{-} $	12.924 461(63) GHz	12.9242(13) GHz	0.3(1.3) MHz	2.2 MHz

TABLE II. Amplitudes of the states with different spin projections. The first arrow corresponds to the projection of the antiproton spin and the second one denotes the projection of the electron spin.

$\overline{\ket{\mathcal{J}, ilde{m}_{\mathcal{J}};\mathcal{F}}}$	$(\uparrow,\uparrow)$	$(\uparrow,\downarrow)$	$(\downarrow,\uparrow)$	$(\downarrow,\downarrow)$
$ 35, 35; 34\frac{1}{2}\rangle$	-0.1187	0.9929	0	0
$ 36, 36; 35\frac{1}{2}\rangle$	1	0	0	0
$ 34, 34; 34\frac{1}{2}\rangle$	0.0201	-0.1178	-0.1178	0.9858
$ 35, 35; 35\frac{1}{2}\rangle$	-0.1170	-0.0140	0.9930	0

 $V_4$  and  $V_5$ , as their coupling constants have more natural interpretations [17].

The potentials  $V_{4-5}$  and  $V_{4+5}$  have exactly the same orbital part and differ only in the spin part (they contain antiprotonic and electronic spin, respectively). We are interested in the states with high orbital number ( $\mathcal{L}=35$ ) and total angular momentum  $\mathcal{J}\approx\mathcal{L}$ . For such states, both spins are either (almost) parallel or antiparallel to  $\mathcal{L}$ . We are considering the transitions  $\nu_{\rm HFS}^\pm$  (see Fig. 2), where spins may flip, but the orbital part does not change. Thus, we can say that each of the potentials  $V_{4\pm5}$  contributes only to the transition where the respective spin flips. To see how the spins  $\mathbf{s}_{\bar{p}}$  and  $\mathbf{s}_{e}$  behave in the transitions  $\nu_{\rm HFS}^\pm$ , we need to expand the four states in question according to Eq. (4). The Clebsh-Gordan coefficients take the simplest form for the states with maximum

projection  $\tilde{m}_{\mathcal{I}}$ . The amplitudes of the states with various spin projections are presented in Table II. The transition  $\nu_{\text{HFS}}^+$  links the first pair of states, while  $\nu_{\text{HFS}}^-$  links the last pair. In the first approximation, both transitions correspond to an electron spin flip—the admixture of the spin flip of the antiproton is suppressed roughly by two orders of magnitude. We see that the expectation value of the potential  $V_{4-5}$  is practically the same for the upper and lower states and, therefore, the transition frequencies  $\nu_{\text{HFS}}^{\pm}$  are practically not affected by this potential, so we are not constraining it. For all other potentials, the electron spin flip causes a sign-change of their expectation values. In the following, we focus on the potentials  $V_2$ ,  $V_3$ ,  $V_{4+5}$ , and  $V_8$ .

For every considered potential  $V_i$ , we introduce the operator  $\mathcal{V}_i$ , defined as  $V_i = f_i^{e\bar{p}} \mathcal{V}_i$ . Then we may estimate the energy shift between states  $|\mathcal{J}_a; \mathcal{F}_a\rangle$  and  $|\mathcal{J}_b; \mathcal{F}_b\rangle$  caused by a  $\mathcal{V}_i$  operator using first-order perturbation theory and the approximate wave functions as follows:

$$\Delta U_{ab,i}(m_0) = \langle \mathcal{J}_a; \mathcal{F}_a | \mathcal{V}_i(m_0) | \mathcal{J}_a; \mathcal{F}_a \rangle - \langle \mathcal{J}_b; \mathcal{F}_b | \mathcal{V}_i(m_0) | \mathcal{J}_b; \mathcal{F}_b \rangle, \quad (11)$$

where  $V_i$  depends on the intermediate boson mass  $m_0$ , as can be seen in Eqs. (6)–(10). For the given values of the  $f_i^{e\bar{p}}$  parameter and the boson mass  $m_0$ , the exotic potential causes a shift of the transition energy equal to  $f_i^{e\bar{p}} \Delta U_{ab,i}(m_0)$ . The maximal discrepancy between theory

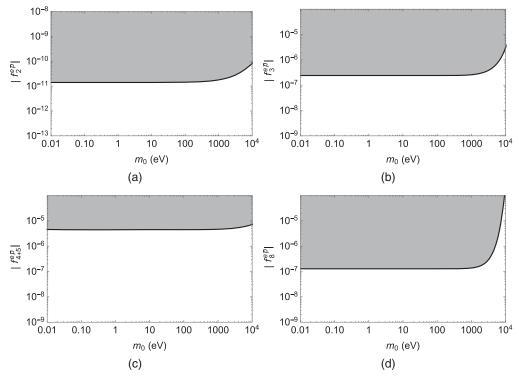


FIG. 3. Constraints (at the 90% confidence level) on the magnitude of the dimensionless coupling constants  $f_i^{e\bar{p}}$  as a function of the boson mass  $m_0$ .

and experiment is equal to  $\Delta E$  (see Table I), so for any value of  $m_0$ , the inequality

$$|f_i^{e\bar{p}}(m_0)\Delta U_{ab\,i}(m_0)| \le \Delta E \tag{12}$$

holds. The constraints on the  $f_i^{ear{p}}$  parameter values can be calculated as

$$|f_i^{e\bar{p}}(m_0)| \le \left| \frac{\Delta E}{\Delta U_{ab,i}(m_0)} \right|. \tag{13}$$

To obtain constraints on  $f_i^{e\bar{p}}$  as a function of  $m_0$ , we perform numerical calculations of  $\Delta U_{ab,i}$  for several  $m_0$  values and then interpolate between them to obtain continuous exclusion plots. We perform this procedure for both transitions,  $\nu_{\rm HFS}^+$  and  $\nu_{\rm HFS}^-$ , and choose the more stringent of the two constraints. The obtained constraints are presented in Fig. 3.

As can be seen in the exclusion plots, for bosons with masses larger than several  $keV/c^2$ , the constraints weaken. This is explained by the fact that our system is less sensitive to interactions mediated by bosons with a Compton wavelength much shorter than the size of the antiprotonic helium atom.

We test our numerically derived constraints by comparing them with results of theoretical estimates (Table III). For these considerations, we use atomic units ( $\hbar = m_e =$ |e|=1) and explore the limit of zero boson mass  $(\lambda \to \infty)$ . The fact that the speed of light is present in potential  $V_2$  as c (in atomic units  $c = 1/\alpha \approx 137$ ), while in the rest of the potentials it comes as  $c^{-1}$  suggests that the constraints on  $V_2$  should be approximately  $\alpha^{-2} \sim 10^4$  times more stringent than on the other potentials. Additionally, due to the spherical symmetry of the electron wave function, for the considered system in potentials  $V_{4\pm 5}$ , only the terms containing derivatives over the antiproton position are relevant [59]. These terms are suppressed by the factor  $m_e/(m_{\bar{p}}+m_e)\approx 0.5\times 10^{-3}$ . Using the virial theorem and taking the potential energy to be ~1 a.u. for an antiproton with  $n \approx 35$ , we may estimate  $\langle \nabla_{\bar{p}} \rangle \sim \sqrt{m_{\bar{p}}}$ , which yields

$$\left\langle \frac{m_e}{m_{\bar{p}} + m_e} \nabla_{\bar{p}} \right\rangle \sim \frac{m_e}{\sqrt{m_{\bar{p}}}} \approx 0.02.$$
 (14)

The other quantities present in the potentials, such as the spins,  $\mathbf{s}_{\bar{p}}$  and  $\mathbf{s}_{e}$ , particle positions,  $\mathbf{r}_{\bar{p}}$  and  $\mathbf{r}_{e}$ , and the

TABLE III. Numerical calculations and order-of-magnitude estimates for constraints on the  $|f_i^{e\bar{p}}|$  parameters in the massless boson limit.

$ f_2 $	$ f_3 $	$ f_{4+5} $	$ f_8 $
$3 \times 10^{-12}$ $1.4 \times 10^{-11}$	$3 \times 10^{-8}$ $2.5 \times 10^{-7}$	$2 \times 10^{-6}$ $4.4 \times 10^{-6}$	$3 \times 10^{-8}$ $1.3 \times 10^{-7}$

differential operator  $\nabla_e$ , can be considered to be of order unity. Comparing the approximate expectation values of the potentials with the value of  $\Delta E \lesssim 2$  MHz  $\approx 3 \times 10^{-10}$  a.u.. from Table I yields the approximate constraints presented in Table III. These constraints are similar to the ones coming from numerical integration. The small differences come from numerical factors, appearing during integration, which are neglected in the estimations. The comparison of constraints obtained in these two ways also yields the conclusion that the use of first-order perturbation theory is justified, as the first order contributions to the energies are not suppressed and therefore are much bigger than the higher order terms.

In conclusion, for the first time, semileptonic spindependent interactions between matter and antimatter have been constrained. This investigation represents an entirely different branch of matter-antimatter comparison tests, complementary to research constraining coupling of exotic bosons to hadronic matter. We obtained the presented results by investigating hypothetical antiproton-electron spin-dependent interactions in antiprotonic helium. Moreover, this analysis provides the first constraints on velocity-dependent spin-dependent matter-antimatter interactions. Our constraints were obtained by comparing theoretical predictions and laboratory results, together with our calculated expectation values of exotic potentials. The current accuracy of the experiment [2] is 20 times higher than the accuracy of the theory [51]. Further improvement in the theory can improve limits obtained in the present work by an order of magnitude.

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<sup>\*</sup>filip.ficek@uj.edu.pl

<sup>[1]</sup> M. Iwasaki, S. N. Nakamura, K. Shigaki, Y. Shimizu, H. Tamura, T. Ishikawa, R. S. Hayano, E. Takada, E. Widmann, H. Outa, M. Aoki, P. Kitching, and T. Yamazaki, Phys. Rev. Lett. **67**, 1246 (1991).

<sup>[2]</sup> T. Pask, D. Barn, A. Dax, R. S. Hayano, M. Hori, D. Horváth, S. Friedreich, B. Juhász, O. Massiczek, N. Ono, A. Sótér, and E. Widmann, Phys. Lett. B 678, 55 (2009).

- [3] H. Nagahama, C. Smorra, S. Sellner, J. Harrington, T. Higuchi, M. J. Borchert, T. Tanaka, M. Besirli, A. Mooser, G. Schneider, K. Blaum, Y. Matsuda, C. Ospelkaus, W. Quint, J. Walz, Y. Yamazaki, and S. Ulmer, Nat. Commun. 8, 14084 (2017).
- [4] C. Smorra, S. Sellner, M. J. Borchert, J. A. Harrington, T. Higuchi, H. Nagahama, T. Tanaka, A. Mooser, G. Schneider, M. Bohman, K. Blaum, Y. Matsuda, C. Ospelkaus, W. Quint, J. Walz, Y. Yamazaki, and S. Ulmer, Nature (London) 550, 371 (2017).
- [5] S. Friedreich, D. Barna, F. Caspers, A. Dax, R. S. Hayano, M.Hori, D. Horváth, B. Juhász, T. Kobayashi, O. Massiczek, A. Sótér, K. Todoroki, E. Widmanna, and J. Zmeskal, Phys. Lett. B 700, 1 (2011).
- [6] M. Hori, H. Aghai-Khozani, A. Sótér, D. Barna, A. Dax, R. Hayano, T. Kobayashi, Y. Murakami, K. Todoroki, H. Yamada, D. Horváth, and L. Venturelli, Science 354, 610 (2016).
- [7] D. Bakalov and V. I. Korobov, Phys. Rev. A 57, 1662 (1998).
- [8] V. I. Korobov, Phys. Rev. A 61, 064503 (2000).
- [9] V. I. Korobov, Phys. Rev. A 73, 022509 (2006).
- [10] V. I. Korobov, Phys. Rev. A 77, 042506 (2008).
- [11] M.-H. Hu, S.-M. Yao, Y. Wang, W. Li, Y.-Y. Gu, and Z.-X. Zhong, Chem. Phys. Lett. **654**, 114 (2016).
- [12] T. Yamazaki, N. Morita, R. S. Hayano, E. Widmann, and J. Eades, Phys. Rep. 366, 183 (2002).
- [13] R. S. Hayano, M. Hori, D. Horvth, and E. Widmann, Rep. Prog. Phys. 70, 1995 (2007).
- [14] M. Bordag, V. M. Mostepanenko, and I. Y. Sokolov, Phys. Lett. A 187, 35 (1994).
- [15] E. J. Salumbides, W. Ubachs, and V. I. Korobov, J. Mol. Spectrosc. 300, 65 (2014).
- [16] J. E. Moody and F. Wilczek, Phys. Rev. D 30, 130 (1984).
- [17] B. A. Dobrescu and I. Mocioiu, J. High Energy Phys. 11 (2006) 005.
- [18] M. S. Safronova, D. Budker, D. DeMille, D. F. Jackson Kimball, A. Derevianko, and C. W. Clark, arXiv:1710 .01833.
- [19] S. Weinberg, Phys. Rev. Lett. 40, 223 (1978).
- [20] F. Wilczek, Phys. Rev. Lett. 40, 279 (1978).
- [21] M. Dine, W. Fischler, and M. Srednicki, Phys. Lett. 104B, 199 (1981).
- [22] M. Shifman, A. Vainshtein, and V. Zakharov, Nucl. Phys. B166, 493 (1980).
- [23] J. Kim, Phys. Rev. Lett. 43, 103 (1979).
- [24] A. R. Zhitnitsky, Yad. Fiz. 31, 1024 (1980) [Sov. J. Nucl. Phys. 31, 529 (1980)].
- [25] F. Wilczek, Phys. Rev. Lett. 49, 1549 (1982).
- [26] G. Gelmini, S. Nussinov, and T. Yanagida, Nucl. Phys. B219, 31 (1983).
- [27] G. Gelmini and M. Roncadelli, Phys. Lett. 99B, 411 (1981).
- [28] Y. Chikashige, R. Mohapatra, and R. Peccei, Phys. Lett. 98B, 265 (1981).
- [29] A. Ansel'm, Pis'ma Zh. Eksp. Teor. Fiz. 36, 46 (1982).
- [30] J. Scherk, Phys. Lett. 88B, 265 (1979).
- [31] D. E. Neville, Phys. Rev. D 21, 2075 (1980).
- [32] D. E. Neville, Phys. Rev. D 25, 573 (1982).

- [33] S. M. Carroll and G. B. Field, Phys. Rev. D 50, 3867 (1994).
- [34] P. Svrcek and E. Witten, J. High Energy Phys. 06 (2006) 051.
- [35] L. B. Okun', J. Exp. Theor. Phys. 56, 502 (1982).
- [36] B. Holdom, Phys. Lett. 166B, 196 (1986).
- [37] B. A. Dobrescu, Phys. Rev. Lett. 94, 151802 (2005).
- [38] C. Bouchiat and C. A. Piketty, Phys. Lett. **128B**, 73 (1983).
- [39] T. Appelquist, B. A. Dobrescu, and A. R. Hopper, Phys. Rev. D 68, 035012 (2003).
- [40] V. A. Dzuba, V. V. Flambaum, and Y. V. Stadnik, Phys. Rev. Lett. 119, 223201 (2017).
- [41] G. Bertone, D. Hooper, and J. Silk, Phys. Rep. 405, 279 (2005).
- [42] A. Friedland, H. Murayama, and M. Perelstein, Phys. Rev. D 67, 043519 (2003).
- [43] V. Flambaum, S. Lambert, and M. Pospelov, Phys. Rev. D 80, 105021 (2009).
- [44] P. W. Graham, D. E. Kaplan, and S. Rajendran, Phys. Rev. Lett. 115, 221801 (2015).
- [45] F. Ficek, D. F. Jackson Kimball, M. G. Kozlov, N. Leefer, S. Pustelny, and D. Budker, Phys. Rev. A 95, 032505 (2017).
- [46] T. M. Leslie, E. Weisman, R. Khatiwada, and J. C. Long, Phys. Rev. D 89, 114022 (2014).
- [47] S. Kotler, R. Ozeri, and D. F. Jackson Kimball, Phys. Rev. Lett. 115, 081801 (2015).
- [48] Y. V. Stadnik, arXiv:1711.03700.
- [49] S. G. Karshenboim, Phys. Rev. D 82, 113013 (2010).
- [50] S. G. Karshenboim and V. V. Flambaum, Phys. Rev. A 84, 064502 (2011).
- [51] V. I. Korobov and D. Bakalov, J. Phys. B 34, L519 (2001).
- [52] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevLett.120.183002 for a derivation of this Hamiltonian.
- [53] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevLett.120.183002 for a derivation of this wave function, which includes Refs. [12,54–56].
- [54] D. J. Griffiths, *Introduction to Quantum Mechanics* (Pearson Education, London, 2005).
- [55] H. A. Bethe and E. E. Salpeter, Quantum Mechanics of Oneand Two-Electron Atoms (Academic Press Inc, New York, 1957)
- [56] J. J. Sakurai, Modern Quantum Mechanics (Addison Wesley, Reading, 1994).
- [57] See Supplemental Material at http://link.aps.org/ supplemental/10.1103/PhysRevLett.120.183002 for a derivation of these parameters and a justification for the use of the variational method in this case.
- [58] The choice of letters denoting sums of angular momenta is inconsistent in the literature, e.g., in Ref. [7]  $J = \mathcal{L} + s_e$  and F is the total angular momentum, while in Ref. [5] the authors also use the letter G. We follow the convention used in Refs. [2,51], using a curly font to avoid possible ambiguity.
- [59] See Supplemental Material at http://link.aps.org/ supplemental/10.1103/PhysRevLett.120.183002 for a formal derivation of this fact.