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Manifestation of the nuclear anapole moment in the M1 transitions in bismuth

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Abstract. Results of the *ab initio* calculation of the parity non-conserving optical rotation caused by the weak charge and the anapole moment of the nucleus are given for the 876 nm and 648 nm transitions in atomic bismuth.

1. Introduction

At present there is good agreement between the atomic experiments on parity non-conservation (PNC) and weak interaction theory. However, there is still no reliable experimental evidence of the existence of the nuclear spin-dependent PNC effects, which were predicted long ago. According to Flambaum and Khriplovich (1980), Flambaum *et al* (1984, 1986) and Bouchiat and Piketty (1991a) the main contribution to these effects arises from the anapole moment of the nucleus, which is caused by the *P*-odd nuclear forces. Therefore, an experimental observation of the nuclear spin-dependent PNC effects in atoms can lead to the discovery of the new electromagnetic moment of a nucleus and will give information about the *P*-odd nuclear forces.

The first calculations of the nuclear spin-dependent *P*-odd transition amplitudes were made by Novikov *et al* (1977) for several heavy atoms (Cs, Tl, Pb and Bi). Later, for the caesium atom more elaborate calculations were made by Kraftmakher (1988), Frantsuzov and Khriplovich (1988) and Blundell *et al* (1990). Quite recently one more semi-empirical calculation for Tl, Pb and Bi was published by Khriplovich (1995). Up to now the only *ab initio* calculation for an atom with more than one valence electron was made for Dy (Dzuba *et al* 1994). This is in contrast with the large number of publications devoted to the *ab initio* calculations of the nuclear spin-independent *P*-odd transition amplitudes. In particular, calculations for bismuth were made by Mårtensson *et al* (1981), Plummer and Grant (1985) and Dzuba *et al* (1989) (for a list of earlier publications see the book by Khriplovich (1991)).

PNC experiments for the 876 nm and 648 nm transitions in bismuth have been carried out in several laboratories (Barkov and Zolotorev 1980, Hollister *et al* 1981, Birich *et al* 1984, Macpherson *et al* 1991, Warrington *et al* 1993). The Oxford group (Macpherson *et al* 1991) has reached an accuracy of 2% for the 876 nm transition and obtained an upper bound for the nuclear spin-dependent *P*-odd transition amplitude which is rather close to the theoretical prediction based on atomic calculations (Novikov *et al* 1977, Khriplovich 1995) and calculations of the nuclear anapole moment (Flambaum and Khriplovich 1980, Flambaum *et al* 1984, 1986, Bouchiat and Piketty 1991a).

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We think that at this point it is time to re-examine the atomic part of the theory. Calculations of Novikov *et al* (1977) as well as the latest calculations of Khriplovich (1995) were based on a semi-empirical model. For the red transition in bismuth the result of this model for the nuclear spin-independent PNC effect does not agree with that of Dzuba *et al* (1989) and with the latest experiment (Warrington *et al* 1993). Note also, that the calculation of Frantsuzov and Khriplovich (1987) for caesium gave results which differ from those of Novikov *et al* (1977) not only in overall scaling, but also in relative values of PNC amplitudes for different hyperfine components of the transition. This is especially important for the interpretation of the experiment as it changes the shape of the expected PNC signal.

In this paper we calculated PNC optical rotation using the configuration interaction (CI) method. Preliminary results of this work were reported in Kozlov and Porsev (1995)

2. PNC amplitudes

The Hamiltonian of the PNC interaction consists of the nuclear spin-independent and nuclear spin-dependent parts:

$$H^{\rm PNC} = H_1^{\rm PNC} + H_2^{\rm PNC} = \frac{G_{\rm F}}{\sqrt{2}}\left(-\frac{Q_{\rm W}}{2}\gamma_5 + \frac{\kappa}{I}\gamma_0\boldsymbol{\gamma}\boldsymbol{I}\right)n(\boldsymbol{r}) \tag{1}$$

where $G_{\rm F} = 2.2225\times10^{-14}$ au is the Fermi constant of the weak interaction, γ_i are the Dirac matrices, $\gamma_5 = \left(\begin{smallmatrix} 0 & -I \\ -I & 0 \end{smallmatrix}\right)$, $\boldsymbol{I}$ is the nuclear spin and $n(\boldsymbol{r})$ is the nuclear density distribution. Atomic units are used throughout the paper.

Dimensionless constants $Q_{\rm W}$ (which is often called the weak charge of the nucleus) and κ characterize the strength of the nuclear spin-independent and nuclear spin-dependent parts, respectively. In the standard model

$$Q_{\rm W} = -N + Z(1 - 4\sin^2\theta_{\rm W}) \tag{2}$$

where $\theta_{\rm W}$ is the Weinberg angle.

The constant κ includes contributions from the anapole moment $\kappa_{\rm a}$ and from the e–N neutral currents $\kappa_{\rm eN}$. One more contribution to the constant κ has been calculated by Flambaum and Khriplovich (1985) and Bouchiat and Piketty (1991b). It comes from the second-order amplitude including the PNC interaction $H_1^{\rm PNC}$ and the magnetic hyperfine interaction $H^{\rm hf}$. This amplitude can be written as the matrix element of an effective operator of the same form as $H_2^{\rm PNC}$ with the constant $\kappa_{\rm eff}$ proportional to the product of the weak charge and the magnetic moment of the nucleus.

Numerical values of the anapole constant $\kappa_{\rm a}$ obtained in Flambaum and Khriplovich (1980), Flambaum *et al* (1984, 1986) differ significantly from those from Bouchiat and Piketty (1991a), but in both cases this constant dominates over two others. For ^{209}Bi ($I = \frac{9}{2}$) all three terms are of the same sign and are of the order of

$$\kappa = \tfrac{10}{11}\kappa_{\rm a} + \kappa_{\rm eN} + \kappa_{\rm eff} \approx 0.3 + 0.05 + 0.08\,. \tag{3}$$

The PNC interactions lead to the admixture of the E1 amplitude to the M1 transition:

$$\mathrm{E1}_{ab}^{\rm PNC} = \sum_n \left(\frac{\langle a|H^{\rm PNC}|n\rangle\langle n|\mathrm{E1}|b\rangle}{E_a - E_n} + \frac{\langle a|\mathrm{E1}|n\rangle\langle n|H^{\rm PNC}|b\rangle}{E_b - E_n}\right) \tag{4}$$

where E_i is the energy of the state i. The sum in this equation runs over the states which satisfy the selection rules for the total electronic angular momentum $\boldsymbol{J}$ and parity. In bismuth we are interested in the M1 transitions between the states of the first odd

configuration $6s^2 6p^3$. For the two lowest transitions (876 and 648 nm) there is no particular even state n dominating this sum.

PNC effects are usually characterized in terms of the parameter

$$R = \mathrm{Im}\left(\frac{\mathrm{E1}_{ab}^{\mathrm{PNC}}}{\mathrm{M1}_{ab}}\right) \tag{5}$$

or, sometimes, in terms of $\mathcal{P} = -2R$.

For the nuclear spin-independent PNC interaction R is independent of the quantum numbers F, F' of the total angular momentum $\boldsymbol{F}$. For the nuclear spin-dependent PNC interaction Novikov *et al* (1977) obtained the following approximate expression:

$$R = \sum_{k=0}^{2} \eta_k (2k+1)^{1/2} \begin{Bmatrix} I & I & 1 \\ J' & J & k \\ F' & F & 1 \end{Bmatrix} \tag{6}$$

where constants η_k depend on the electronic degrees of freedom. This formula is valid if one neglects the differences in the energy denominators in (4) for the intermediate states which belong to the same configuration. This approximation is good when the distances between the states of any important intermediate configuration are much smaller than the distances between this configuration and the states a and b. In fact, for bismuth this is not true. There are several important configurations, such as the configuration $6s6p^4$, which are very wide on the energy scale and the corresponding denominators can differ by a factor of two. For this reason we used the initial expression (4) without averaging over the energy denominators.

3. Method

In this paper we used the relativistic CI method with the Hartree–Fock–Dirac (HFD) core, the last core shell being 5s. Twenty-one electrons from the 5p, 5d, 6s and 6p shells were included in the CI procedure. The sum (4) was found by solving the inhomogeneous equation in the CI space. For the HFD and CI calculations we used significantly modified versions of the codes written by Bratcev *et al* (1977) and Kotochigova and Tupitsin (1987).

Basis set. Unlike the perturbation theory, where the eigenfunctions of the unperturbed Hamiltonian should be used, the CI method allows us to choose the basis set for better convergence and convenience (the relativistic CI basis functions should belong to the positive-energy subspace Λ^+). It is still reasonable to use HFD orbitals for the core and valence shells to have a good zero approximation. In particular, orbitals 1s...6p were obtained from the HFD equations for the configuration $6s^2 6p^3$. After that, these orbitals were frozen and the 7s orbital was found from the HFD equation for the configuration $6s^2 6p^2 7s$.

The usage of the HFD orbitals for the virtual shells is inefficient for two reasons. First, mixing of the Rydberg states to the CI wavefunction is suppressed because of their localization at large distances. Their contribution to the PNC effects is additionally suppressed due to the small overlap with the nucleus†. Second, it is technically inconvenient to work with the functions from the continuum.

The more adequate basis set should consist of the orbitals, which are mainly localized at the same distances from the origin as the valence ones. For our purposes it is also very

† For example, the contributions to the PNC amplitude between the hyperfine levels of the ground state of hydrogen look as follows: 2p state—35%, other discrete states—12%, continuum—53% (Gorshkov *et al* 1988).

important that these orbitals have proper behaviour at the origin (i.e. $\sim r^{\gamma_j}$ for the point-like nucleus). To satisfy these conditions one can take any linear combination of the HFD orbitals of the proper symmetry plus function ϕ, which is rapidly tending to zero both at the origin and at infinity. One can choose ϕ, for example, as r times the previous orbital of the same symmetry.

In fact, this procedure was used only for the large component f_{nlj} of the radial Dirac bispinor. The small component g_{nlj} was found from the kinetic balance condition: $g_{nlj} = \frac{1}{2mc}(\boldsymbol{\sigma p}) f_{nlj}$, which is equivalent to the projection on the subspace Λ^+. The radial function $\binom{f}{g}$ was orthonormalized with respect to the functions of the same symmetry, that fixed coefficients of the linear combination of the HFD functions. Similar basis sets were used in the non-relativistic atomic calculations previously (see, for example, Bogdanovich 1988, Shirley and Martin 1993). In the relativistic atomic calculations non-HFD virtual orbitals were used by Dinov *et al* (1994) (see also references therein).

Finally, our basis set consisted of 5p, 5d, 6s, 6p and 7s HFD orbitals plus 8–11s, 7–9p, 6, 7d and 5f virtual orbitals. Almost all virtual functions corresponded to the positive Hartree–Fock energy. Thus, they accounted for both Rydberg and continuum parts of the HFD spectrum.

CI space. All odd configurations used in our calculations were produced from the configurations $6s^2 6p^3$ and $6p^5$. No triple excitations were included. To account for the core polarization effects CI included all single and part of the double excitations from the 5p and 5d shells.

Even configurations were also produced from the same two odd configurations. The most important for the sum (4) were the following three sets of even configurations:

$$6s^2 6p^2 n\mathrm{s} \quad n = 7\text{–}11 \qquad 6s6p^4, 6s6p^3 n\mathrm{p} \quad n = 7\text{–}9 \qquad 6s^2 6p^2 n\mathrm{d} \quad n = 6, 7\,. \tag{7}$$

Note that the third set of configurations corresponds to the 6p–nd transitions and thus does not contribute directly to the PNC amplitude. Nevertheless, it was very important because it strongly interacts with the two other sets. It is not surprising because configurations $6s^2 6p^2 n$d are connected to configurations $6s^2 6p^2 n$s and $6s6p^3 n$p by the large dipole–dipole transitions and lie in the same spectral region.

Inhomogeneous equation. The sum (4) can be rewritten in terms of the solution of the inhomogeneous equation:

$$\mathrm{E1}_{ab}^{\mathrm{PNC}} = \langle \phi_a | \mathrm{E1} | b \rangle + \langle a | \mathrm{E1} | \phi_b \rangle \tag{8}$$

$$(E_i - H)|\phi_i\rangle = H^{\mathrm{PNC}}|i\rangle \tag{9}$$

or, similarly

$$\mathrm{E1}_{ab}^{\mathrm{PNC}} = \langle \chi_a | H^{\mathrm{PNC}} | b \rangle + \langle a | H^{\mathrm{PNC}} | \chi_b \rangle \tag{10}$$

$$(E_i + \omega - H)|\chi_i\rangle = \mathrm{E1}|i\rangle \tag{11}$$

where $\omega = E_f - E_i$ is the transition frequency. We solved both (8), (9) and (10), (11) to check that the amplitude $\mathrm{E1}_{ab}^{\mathrm{PNC}}$ was the same.

We used L- and V-gauges for the operator E1. Results obtained with the former appeared to be more stable, while for the latter large cancellations took place. That is typical for the low-frequency transitions. In particular, two matrix elements (8) were of different sign for the V-gauge and of the same sign for the L-gauge.

4. Results and discussion

To check the accuracy of the method we calculated the energies, g-factors and the hyperfine constants A and B for all five levels of the configuration $6s^2 6p^3$. Tables 1–3 show our results in comparison with experimental data and recent calculations of Dzuba *et al* (1989, 1991). Table 1 also includes the energies of five lower even levels. It is seen that there is good agreement with the experiment for the energies of the levels of both parities.

Table 1. Energies (in cm^{-1}) of the five odd and five even low-lying states of bismuth.

		Experiment[a]	MBPT[b]	This work
Odd	$^4S^o_{3/2}$	0	0	0
	$^2D^o_{3/2}$	11 419.0	11 672	11 521
	$^2D^o_{5/2}$	15 437.7	15 593	15 969
	$^2P^o_{1/2}$	21 661.0	21 806	22 222
	$^2P^o_{3/2}$	33 164.8	33 337	33 185
Even	$^4P_{1/2}$	32 588.2	—	32 823
	$^4P_{3/2}$	44 865.1	—	44 418
	$^2P_{1/2}$	45 915.6	—	45 814
	$^4P_{5/2}$	48 498.9	—	48 940
	$^2P_{3/2}$	49 456.6	—	49 599

[a] Moore (1958).
[b] Dzuba *et al* (1989).

Table 2. g-factors of the states of configuration $6s^2 6p^3$ with $J = \frac{3}{2}$.

	$^4S^o_{3/2}$	$^2D^o_{3/2}$	$^2P^o_{3/2}$
Experiment[a]	1.6433	—	1.2608
MBPT (SO)[b]	1.5907	1.2884	1.2645
MBPT (HO)[c]	1.6445	1.2321	1.2594
CI (small)[d]	1.7433	1.1560	1.2340
CI (large)[e]	1.7052	1.1789	1.2495

[a] Title and Smith (1960), Landman and Lurio (1970).
[b] Second order in the residual e–e interaction (Dzuba *et al* 1989).
[c] Semi-empirical higher order corrections included (Dzuba *et al* 1989).
[d] CI includes five relativistic configurations.
[e] CI includes 355 relativistic configurations.

Table 2 shows that the accuracy of our calculation for the intermediate coupling coefficients†, which determine g-factors, is comparable to the accuracy of the second-order many-body perturbation theory (MBPT) but is not quite satisfactory. The hyperfine structure for the two lower levels appears to also be very sensitive to the intermediate coupling coefficients. Note that in the zeroth approximation even the sign of the constant A for the ground state is wrong (see table 3). For most of the other hyperfine parameters our accuracy is about 10%.

† By the intermediate coupling coefficients we mean the mixing coefficients of three relativistic configurations which correspond to the configuration $6s^2 6p^3$.

Table 3. Hyperfine constants A and B (MHz).

	$^4S^o_{3/2}$		$^2D^o_{3/2}$		$^2D^o_{5/2}$		$^2P^o_{1/2}$	$^2P^o_{3/2}$	
	A	B	A	B	A	B	A	A	B
Experiment[a]	−447	−305	−1230	−653	2503	14	11 268	491	979
CI (small)[b]	92	−171	−550	−910	2809	0	10 843	849	1090
CI (large)[b]	−667	−178	−993	−729	2340	29	10 606	450	1009

[a] Khriplovich (1991, see references therein).
[b] The quadrupole moment of the ^{209}Bi nucleus was taken to be equal to -50×10^{-24} cm^2 (Pyykkö and Li 1992).

The M1 transition amplitudes are directly involved in the calculation of the PNC factor R (see equation (5)). Results for these amplitudes are listed in table 4. Experimentally only the squared ratio of the first two amplitudes has been measured.

Table 4. M1 transition amplitudes for the states of configuration $6s^26p^3$: $^4S^o_{3/2}$–$^2D^o_{3/2}$ (876 nm), $^4S^o_{3/2}$–$^2D^o_{5/2}$ (648 nm), $^4S^o_{3/2}$–$^2P^o_{1/2}$ (462 nm), $^4S^o_{3/2}$–$^2P^o_{3/2}$ (301 nm). Absolute values for the reduced matrix elements in Bohr magnetons are given.

	876 nm	648 nm	462 nm	301 nm	X[a]
Experiment[b]	—	—	—	—	8.75
MBPT (SO)[c]	1.747	0.615	0.625	0.166	8.07
MBPT (HO)[c]	1.695	0.563	0.590	0.199	9.06
CI (small)	1.521	0.450	0.557	0.275	11.42
CI (large)	1.618	0.510	0.573	0.251	10.07

[a] Parameter X is equal to the squared ratio of the first and second amplitudes.
[b] Macpherson *et al* (1992).
[c] Dzuba *et al* (1989).

We also calculated the lifetimes of the two lowest even states to check the accuracy of the E1 transition amplitudes. In the L-gauge they appeared to be 5.3 ns for the $^4P_{1/2}$ state and 5.1 ns for the $^4P_{3/2}$ state while experiment gives 4.6 ± 0.4 ns and 6 ± 2 ns, respectively (Radtcig and Smirnov 1986).

Summing up all these data we conclude that in spite of some disagreement in the intermediate coupling coefficients, our calculations reproduce most of the experimental features of the lower levels of bismuth. Though the total contribution of the higher even levels to the sum (4) is large, changes in particular terms are not so significant and do not influence the final accuracy.

The calculation procedure for the PNC amplitude $E1^{PNC}$ has been described in the previous section. The $E1^{PNC}$ and M1 amplitudes were used to evaluate R. The results of the calculations in the L-gauge are presented in table 5. Notations small–small, large–small, and large–large are used to distinguish between small and large CI for odd and even states correspondingly. Small CI for the odd states included configurations $6s^26p^3$ and $6p^5$. As it follows from tables 2–4 this is a very rough approximation for the wavefunctions. Small CI for the even states was restricted to the configurations (7). One can see that for the first two transitions there was a 40% difference between the small–small and large–large cases, while for the third transition it was more than 60%. In contrast, the difference between L- and V-gauges decreased from 22% and 18% for the 876 and 648 nm transitions to 11% for the 462 nm one. This corresponded to the smaller cancellations for the higher

frequency transitions in the *V*-gauge. We estimate the accuracy of our calculations of the $E1^{PNC}$ in the *L*-gauge to be better than 10% for the first transition, between 10% and 15% for the second one and up to 20% for the third (the accuracy of the *V*-gauge is approximately two times worse). Within this uncertainty our results agree with the latest experiments of Macpherson *et al* (1991) and Warrington *et al* (1993).

Table 5. Weak charge contribution to PNC in the M1 transitions between the states of configuration $6s^26p^3$.

	$R \times 10^7/(-N/Q_W)$		
Transition	876 nm	648 nm	462 nm
Experiment[a]	-1.012 ± 0.020	-0.98 ± 0.09	—
MBPT [b]	-1.10 ± 0.13	-0.75 ± 0.5	—
CI[c]			
small–small	-1.40	-1.21	-2.97
large–small	-0.91	-0.91	-1.55
large–large	-1.02	-0.94	-1.79

[a] Macpherson *et al* (1991), Warrington *et al* (1993).
[b] Dzuba *et al* (1989).
[c] Small, and large correspond to small and large CI for the odd and even states, respectively. Details are given in the text.

The accuracy for the 648 nm transition deserves a more detailed discussion. Dzuba *et al* (1989) pointed out that in the MBPT approach there are large cancellations for this transition which result in the large uncertainty in R. We also had several large contributions to the sum (4), but there were no large cancellations. Moreover, results appeared to be rather stable against the changes in the calculation procedure. However, it was very important that CI for the even levels included all three sets of configurations (7). Strong mixing between configurations $6s^26p^2nd$ and the two other sets of configurations significantly changed the R parameter for the 648 nm transition. In terms of the MBPT large configuration mixing means that higher order terms are important. This can explain the instability of the MBPT results for the 648 nm transition.

The main source of the uncertainty in the present calculation is associated with the excitations from the closed shells. Indeed, the dimension of the problem for the complete CI depends exponentially on the number of the valence particles. For this reason we included only the most important excitations from the closed shells.

Our final results for the PNC parameters R are as follows:

$$\text{876 nm line:} \quad R = -(1.02 \pm 0.15) \times 10^{-7}\left(\frac{-N}{Q_W}\right) \tag{12}$$

$$\text{648 nm line:} \quad R = -(0.94 \pm 0.15) \times 10^{-7}\left(\frac{-N}{Q_W}\right) \tag{13}$$

$$\text{462 nm line:} \quad R = -(1.79 \pm 0.35) \times 10^{-7}\left(\frac{-N}{Q_W}\right). \tag{14}$$

The nuclear spin-dependent PNC amplitudes were calculated within the same technique. The differences of the results for the small and large CI were similar to those for the nuclear spin-independent amplitudes. In table 6 only the final values for the *L*-gauge are listed.

Operators H_1^{PNC} and H_2^{PNC} are very similar. The main difference is that the former is a scalar, while the latter is a vector in the electronic subspace. That increases the number

Table 6. Nuclear spin-dependent PNC admixture to the M1 transitions $^4S^o_{3/2}$–$^2D^o_{3/2}$ (876 nm) and $^4S^o_{3/2}$–$^2D^o_{5/2}$ (648 nm).

		$R \times 10^8/\kappa$					
		876 nm transition			648 nm transition		
F	F'	SE[a]	SE[b]	CI[c]	SE[a]	SE[b]	CI[c]
3	2	—	—	—	0.56	0.82	0.81
3	3	−0.08	−0.11	−0.12	0.82	1.14	1.15
3	4	−0.01	−0.01	−0.04	1.18	1.58	1.60
4	3	−0.21	−0.30	−0.26	−0.04	0.03	0.01
4	4	0.98	1.42	1.25	0.32	0.46	0.46
4	5	0.11	0.17	0.12	0.76	1.00	1.02
5	4	−0.13	−0.19	−0.15	−0.76	−0.94	−0.97
5	5	−0.13	−0.19	−0.17	−0.32	−0.40	−0.41
5	6	0.25	0.36	0.31	0.22	0.25	0.26
6	5	−0.04	−0.06	−0.02	−1.61	−2.08	−2.13
6	6	0.07	0.09	0.10	−1.08	−1.43	−1.45
6	7	—	—	—	−0.46	−0.67	−0.66

[a] Semi-empirical calculation of Novikov *et al* (1978).
[b] Semi-empirical calculation of Khriplovich (1995).
[c] This work.

of terms in the sum (4) and makes calculations slightly more difficult. For this reason we estimate the accuracy of these calculations to be about 20% for both transitions.

In general there is good agreement between our results and those of Novikov *et al* (1977) and Khriplovich (1995). It is seen, that the simplification (6) used by Novikov *et al* works quite well for the first transition, but slightly changes the F-dependence for the second. Surprisingly, there is a very good agreement for the 648 nm transition with Khriplovich (1995). Note, that for this transition the semi-empirical calculation significantly overestimates the nuclear-spin-independent PNC amplitude.

It is also important to examine the resultant PNC signal from all hyperfine components of the M1 transition. That can be readily done with the help of the data from table 6 and the experimental line width parameters $\Gamma_L = 350$ MHz and $\Gamma_D = 650$ MHz (Macpherson *et al* 1992).

For these parameters all three calculations give almost identical signal shapes, the only difference being in the overall scaling.

Finally, we would like to mention that the nuclear spin-dependent and nuclear spin-independent PNC signals are almost orthogonal to each other for the 876 nm transition, but not for the 648 nm transition. For this reason it can be more difficult to separate the nuclear spin-dependent signal in the second case.

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