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Precision Spectroscopy of the ^{207}Pb^{19}F molecule: implications for measurement of $P-$odd and $T-$odd effects.

Lukas D. Alphei, Jens-Uwe Grabow
Gottfried-Wilhelm-Leibniz-Universität, Institut für Physikalische Chemie & Elektrochemie, D-30167 Hannover, Germany

A.N. Petrov
Petersburg Nuclear Physics Institute, Gatchina, Leningrad district 188300, Russia,
Institute of Physics, Saint Petersburg State University, Saint Petersburg, Petrovoretz 198904, Russia

Richard Mawhorter, Benjamin Murphy, Alexander Baum
Department of Physics and Astronomy, Pomona College, Claremont CA 91711-6359

Trevor J. Sears
Chemistry Department, Brookhaven National Laboratory, Upton, NY 11973-5000

T. Zh.Yang, P.M Rupasinghe, C.P. McRaven, N.E. Shafer-Ray
Homer L. Dodge Department of Physics and Astronomy, University of Oklahoma, Norman OK, USA

Here we report for the first time precision microwave spectroscopy of pure rotational transitions of the ^{207}Pb^{19}F isotopologue. We use these data to make predictions of of the sensitivity of the molecule to P-odd, T-even and P-odd, T-odd effects.

It has long been established that the spectroscopy of heavy paramagnetic diatomic molecules is fertile ground for the study of new physics. Such molecules enhance interactions that exhibit violation of discrete symmetries. The extent of this symmetry violation may in turn shed light on such problems as that of baryogenesis and help to differentiate between the Standard Model and alternate theories of matter. One reason that heavy paramagnetic molecules enhance the effect of discrete symmetry violation is that such molecules have closely spaced levels of opposite parity. This close spacing allows states of mixed parity to be accessible at laboratory-strength electromagnetic fields. In addition to this accessibility, nuclear interactions grow in strength with mass, enhancing both nuclear effects that are odd upon the inversion of spatial symmetry (P-odd) and/or reversal of time (T-odd.) Finally, the fact that the electronic orbit of an unpaired electron is highly relativistic provides a linear sensitivity to an e-EDM, despite the fact that (classically) the average electric field must be zero[1–3].

The current limit on $e-$EDM is the result of the probe of the Tl atom[4, 5]. In recent years, many groups have reported experiments that exploit the sensitivity of heavy diatomic molecules to an $e-$EDM. These efforts include probes of PbO*[6–9],YbF[10], HfF$^+$[11], WC[12], and ThO*[13] and our own work with the PbF molecule[14]. Although these efforts have yet to yield a significant limit, continual advances in experimental methods may soon change this situation. In light of ongoing efforts, a judgement the relative effectiveness of the varying molecular systems and techniques is premature. We can, however, point to advantages of the PbF molecule that have led us to study this system[14–21]. The PbF molecule has an intrinsic sensitivity to an $e-$EDM that is roughly equal to that of YbF, PbO*, and HfF$^+$ and roughly 1/3 the sensitivity of WC and ThO*. The ground state of the PbF molecule is sensitive to the $e-$EDM so that the coherence time is not lifetime limited as it is for the PbO* (0.1 ms [6]) and ThO*(1 ms [22]) systems. As a $^2\Pi_{1/2}$ molecule, the contribution to the magnetic moment of PbF due to the electronic orbital angular momentum roughly cancels the contribution due to the unpaired electron spin angular momentum. For this reason, the $g-$factor of a polarized PbF molecule is small ($g \approx 0.04$), greatly easing the requirements on the control of magnetic fields needed to carry out a precise $e-$EDM experiment. We note that others are studying $^3\Delta_1$ states of the molecules WC[12], ThO*[23], and HfF$^+$[11] that exhibit similarly small $g-$factors.

A possible advantage of the PbF molecule is that its complex hyperfine structure allows for nearly-degenerate states of opposite parity. Here we present an analysis of spectra and derive molecular constants that describe the hyperfine split spin-rotational levels of the ground state of PbF. This knowledge is critical to the design and interpretation of ongoing efforts to use PbF in an $e-$EDM measurement. The sensitivity of the PbF molecule to an $e-$EDM has motivated detailed optical spectroscopy[14, 18, 19, 21, 24]. This optical spectroscopy led to the prediction of energy levels to an accuracy of approximately 10 MHz which in turn allowed for efficient production of the pure rotational microwave spectra of the ^{207}Pb^{19}F isotopologue that we present here. These measurements are carried out using a Balle-Flygare-type[25] Fourier transform microwave (FT-MW) spectrometer at the Gottfried-Wilhelm-Leibniz-Universität Hannover. This apparatus uses a coaxial arrangement of the supersonic expansion and resonator axis (“Coaxially Oriented Beam-Resonator Arrangement”, COBRA)[26, 27]. The diatomic compound is synthesized in situ using laser ablation[28, 29] (Nd:YAG, 1064 nm, 250 mJ, 20 Hz) of a rotating, elementary Pb rod in the presence of highly diluted SF_6 entrained in a noble carrier gas. Neon (4.5 – 6 bar absolute) was used as the carrier gas. Specifically, we present an interpretation of the measurement of hyperfine split pure rotational spectra of the ^{207}Pb^{19}F molecule (31 transitions) that is supplemented by pure rotational spectra of ^{206}Pb^{19}F (13 transitions) and ^{208}Pb^{19}F (19 transitions.) These transitions range in frequency from 3.2 GHz to 26.1 GHz with an absolute accuracy of 300 Hz (typical) and involve rotational quantum numbers from $J = 1/2$ to $J = 5/2$. In addition to these field free transitions, Zeeman splittings were measured in order to determine molecular-frame $g-$factors that characterize the Zeeman effect. Finally, an optical study of the Stark effect was carried out at the University of Oklahoma to determine the permanent dipole moment of the molecule. Details of these measurements and our analysis will be given in a future publication.

To fit the data with a variance that matches the experimental accuracy, the energy levels of the ^{207}Pb^{19}F molecule are modeled with an effective spin-rotational Hamiltonian $\mathbf{H}_{\mathrm{SR}} = \mathbf{H}_{rot} + \mathbf{H}_0 + \mathbf{H}_1 + \mathbf{H}_{ext}$. The interaction $\mathbf{H}_{rot}$ describes the spin-rotational motion of the molecule, neglecting all nuclear spin. The interaction $\mathbf{H}_0$ describes the hyperfine structure that results from the Frosch and Foley picture[30] of the interaction of nuclear spin with an electron in a specified quantum orbit. The interaction $\mathbf{H}_1$ gives small high-order corrections to the hyperfine structure. Finally, $\mathbf{H}_{ext}$ describes the interaction of the molecule with external fields.

The rotational Hamiltonian $\mathbf{H}_{rot}$ of a $^2\Pi$ molecule is best described in terms of a Hamiltonian that mixes both the $^2\Pi_{1/2}$ and $^2\Pi_{3/2}$ states. The procedure for determining the rotational energies from this Hamiltonian is described in many sources (see, for example, [31, 32].) In our fit to our observed spectra of ^{207}Pb^{19}F, we determine an energy level structure characterized by the parameters (in MHz) $A = 248,117,014(2)$, $B = 6,917.9108(10)$, $p = -4,145.2304(3)$, $A_D = 157.667(30)$, $p_D = -0.0031(4)$, and $D = 0.00553(2)$. The constant $A_H = 0.0000899(3)$ MHz has been determined previously by precision measurement of higher rotational states by Ziebarth and coworkers[33]. The remaining interactions $\mathbf{H}_0$, $\mathbf{H}_1$, and $\mathbf{H}_{ext}$ are given by

$$\mathbf{H}_0 = \mathbf{I}'_1 \cdot \widehat{A}_1 \cdot \mathbf{S}' + \mathbf{I}'_2 \cdot \widehat{A}_2 \cdot \mathbf{S}' \tag{1}$$

$$\begin{aligned} \mathbf{H}_1 = (c_1\ \mathbf{I}_1 \cdot \mathbf{J} + c_2\ \mathbf{I}_2 \cdot \mathbf{J}) \\ -t_0\ \left[3\ (\mathbf{I}'_1 \cdot \mathbf{n}')(\mathbf{I}'_2 \cdot \mathbf{n}') - (\mathbf{I}_1 \cdot \mathbf{I}_2)\right] \\ +(d_{C1}\ \mathbf{I}_1 \cdot \mathbf{J} + d_{C2}\ \mathbf{I}_2 \cdot \mathbf{J})\ \ (\mathbf{J}' \cdot \mathbf{S}') \end{aligned} \tag{2}$$

$$\begin{aligned} \mathbf{H}_{ext} = \mu_B\ \mathbf{B}' \cdot \widehat{G} \cdot \mathbf{S}' + D\ \mathbf{E} \cdot \mathbf{n}' \\ -g_1\ \mu_N \mathbf{B} \cdot \mathbf{I}_1 - g_2\ \mu_N \mathbf{B} \cdot \mathbf{I}_2 \end{aligned} \tag{3}$$

Here the subscripts 1 and 2 refer to nuclear spin of the fluorine and lead, respectively. Primes indicate quantities in the body-fixed frame of the molecule whereas quantities without primes are taken with respect to the laboratory frame. The vector $\mathbf{n}'$ gives the direction of the internuclear axis. The angular momentum operators $\mathbf{I}_1$, $\mathbf{I}_2$,and $\mathbf{J}$ represent the angular momenta of the fluorine and lead nuclei, and the total angular momentum of the molecule neglecting nuclear spin. The tensor contractions $\mathbf{I}' \cdot \widehat{A} \cdot \mathbf{S}' = A_{||}\mathbf{I}_0^{1\prime}\mathbf{S}_0^{1\prime} - A_{\perp}(\mathbf{I}_1^{1\prime}\mathbf{S}_{-1}^{1\prime} + \mathbf{I}_{-1}^{1\prime}\mathbf{S}_1^{1\prime})$ and $\mathbf{B}' \cdot \widehat{G} \cdot \mathbf{S}' = G_{||}\mathbf{B}_0^{1\prime}\mathbf{S}_0^{1\prime} - G_{\perp}(\mathbf{B}_1^{1\prime}\mathbf{S}_{-1}^{1\prime} + \mathbf{B}_{-1}^{1\prime}\mathbf{S}_1^{1\prime})$ are determined by the hyperfine parameters $A_{||}$, $A_{\perp}$ and body-fixed $g-$factors $G_{||}$ and $G_{\perp}$.

Nine nuclear interaction parameters occur in the field-free Hamiltonians $\mathbf{H}_0$ and $\mathbf{H}_1$ required to describe our spectra to experimental accuracy. The four parameters $A_{||1} = 409.906(1)$ MHz, $A_{\perp 1} = 255.991(1)$ MHz, $A_{||2} = 10,146.6733(9)$ MHz, and $A_{\perp 2} = -7,264.0388(4)$ MHz are determined by weighted combinations of Frosch and Foley parameters a, b, c and d, as described elsewhere[21]. The two parameters $c_1 = 0.0093(3)$ MHz and $c_2 = 0.0368(1)$ MHz describe the interaction of the nuclear magnetic moments with the magnetic field of nuclear rotation (see ref. [32], page 535.) The spectra are also sensitive to the parameter $t_o = 0.0022(6)$ MHz describing the nuclear bipolar (magnetic dipole-dipole) interaction (see ref. [32] page 558.) Finally, the precision of the ^{207}Pb^{19}F spectra enabled sensitivity to an Ω-doublet dependence to the nuclear-rotational interaction parameterized by $d_{C1} = 0.00056(10)$ MHz and $d_{C2} = -0.007(2)$ MHz. The sensitivity of the ^{207}Pb^{19}F molecule to external magnetic fields is described by body-fixed $g-$factors $G_{||} = -0.38(2)$ and $G_{\perp} = 0.12(1)$ which are determined by analyzing Zeeman splittings. In the optical measurement at the University of Oklahoma, the electric dipole moment of ^{208}Pb^{19}F is measured to be $D = 3.5(3)$ Debye. The resulting energy level structure of the $J = 1/2$ and $J = 3/2$ states of ^{207}Pb^{19}F is shown in Fig. 1. The most striking feature of this structure is closely spaced levels of opposite parity in the $J = 1/2$ manifold. This coincidental near degeneracy, first observed in lower-resolution optical spectra[19], is measured here to be 266.285 MHz between the $F = 1/2$, $p_s = -1$, and $F = 1/2$, $p_s = +1$ states. This near proximity of the states of opposite parity allows for states of mixed parity at electric and magnetic fields that are roughly a factor of five smaller than that required for the even isotopologues of PbF.

The sensitivity of the PbF molecule to P- and T-odd effects has been quantified in several research publications[15, 16, 22, 34–36]. The most recent example is a detailed electronic structure calculation that determines the sensitivity parameters W_P, $W_{P,T}$ and W_d for the ^{207}Pb^{19}F molecule[36]. In this work the calculated parameter $W_P = -0.99$ kHz gives the sensitivity to the P-odd, T-even anapole moment of the ^{207}Pb nucleus. The parameter $W_{P,T} = 75$ kHz gives the sensitivity of the molecule to a possible P-odd, T-odd anapole moment. Finally the parameter $W_d = 16$ mHz/(10^{-27}e·cm) gives the sensitivity to an $e-$EDM (which, for ^{207}Pb^{19}F, is indistinguishable from the $W_{P,T}\ \kappa_{P,T}$ interaction.) These parameters are incorporated into the spin-rotational

Hamiltonian with the addition of the interaction

$$\mathbf{H}_{PT} = (W_p\ \kappa_p)\mathbf{n} \times \mathbf{S}'\cdot\mathbf{I}_2 + (W_{P,T}\ \kappa_{P,T} + W_d d_e)\mathbf{S}'\cdot\mathbf{n}. \quad (4)$$

Here $\mathbf{S}'$ and $\mathbf{I}_2$ are the (P-even, T-odd) effective-electronic- and ^{207}Pb-nuclear- spin operators, and $\mathbf{n}$ is the (P-odd, T-even) internuclear axis. Finally κ_p and $\kappa_{P,T}$ are unitless anapole constants[37, 38] for the ^{207}Pb nucleus[37]. The combination of our precisely measured P-even, T-even parameters and calculated sensitivity parameters allows for quantitative predictions of the dependence of ^{207}Pb^{19}F spectra to anapole moments and a possible electron electric dipole moment.

P-odd, T-even effects have been measured in Cs[39]. This atomic measurement probes the influence of parity violation on the transition rate of an otherwise forbidden dipole transition. It is not clear that a molecular probe of similar P-odd, T-even moments would provide more sensitivity than these atomic measurements. It is nevertheless instructive to contrast the inclusion of P-odd, T-even moments in the spin rotational Hamiltonian to inclusion of P-odd, T-odd moments. A signature of the P-odd, T-even molecular interaction $(W_p\ \kappa_p)\ \mathbf{n} \times \mathbf{S}' \cdot \mathbf{I}_2$ is an avoided crossing in the Zeeman spectroscopy of two states with opposite parity. The first such crossing in the ^{207}Pb^{19}F molecule occurs for two $M_F = 1/2$ states that cross at a magnetic field of 823 ± 40 gauss, with an error determined by uncertainty in the measured factors $G_{||}$ and $G_{\perp}$. To illustrate this crossing, we assume an anapole effect $W_p\ \kappa_p = -119$ Hz, as expected from the anapole moment of ^{207}Pb predicted by Flambaum and Khriplovich[37], $\kappa_p = -0.12$. This strength interaction leads to the avoided crossing illustrated in Fig. 1. Measurement of such a splitting is well within the reach of the current measurement. There remain the dual difficulties of creating a very uniform magnetic field and the difficulty of avoiding systematic errors due to stray electric fields. For the case illustrated in Fig. 2, a 68 μvolt/cm electric field directed along the axis of the magnetic field would create an avoided crossing that is indistinguishable from that of the anapole moment. For this reason, it is not immediately apparent that direct observation of this avoided crossing can compete with atomic experiments. Experimental techniques have been proposed that may overcome these issues[40].

A signature of the combined P-odd T-odd interaction $(W_{P,T}\ \kappa_{P,T} + W_d\ d_e)\mathbf{S}'\cdot\mathbf{n}$ is a difference in the energy of two states of the molecule that are identical in every way but the sign of the projection of total angular momentum M_F onto the axis of a pure electric field. Figure 2 gives the energy difference between $\pm M_F$ states of the PbF molecule in units of $W_{P,T}\ \kappa_{P,T} + W_d\ d_e$. The $M_F = \pm 3/2$ system shows the greatest sensitivity to this P-odd, T-odd effect, but the $M_F = \pm 1/2$ system is sensitive at lower values of electric field.

Although we have noted the sensitivity of the ^{207}Pb^{19}F molecule to P-odd, T-even effects, the use of the molecule as a probe of the P-odd, T-odd $e-$EDM has motivated this work. Here we have presented Zeeman microwave spectroscopy with a linewidth that is approximately equal to that reported in the Hinds' group study of YbF[10]. While both PbF and YbF have similar sensitivities to an e-EDM, the g-factor of PbF is 20 times smaller than that of the YbF molecule. We also stress that, as for the case of YbF and WC, the *ground* electronic state of the PbF molecule is sensitive to an $e-$EDM. This final feature allows one to contemplate extremely long coherence times[20] and is a feature not shared by the HfF^{+}[11], PbO* [6–9, 13, 41–43] and ThO* [23, 44] systems currently under investigation.

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FIGURE CAPTIONS

FIGURE 1: Left hand side: Hyperfine energy level structure of the ground electronic (X_1) and rotational ($J = 1/2$) state of the ^{207}Pb^{19}F molecule as a function of F(total angular momentum) and p_s(parity). Two closely spaced $F = 1/2$ states of opposite parity are shown by thick lines. Right hand side: Anapole-induced avoided crossing of the $M_F = 1/2$ sublevels of the closely spaced $M_F = 1/2$ states. This crossing is predicted to occur at a field of 823 ± 40 gauss.

FIGURE 2: P- and T- odd dependent splitting between $\pm M_F$ pairs of hyperfine states of the ground rotational $J = 1/2$ state of ^{207}Pb^{19}F. The thick solid line corresponds to the $|M| = 3/2$lower-lying Ω-doublet states whereas the thin solid line corresponds to the higher lying $|M_F| = 3/2$ states. The dotted lines correspond to $|M_F| = 1/2$ sublevels. For the case that $\kappa_{P,T}$ and $d_e = 1.6 \times 10^{-27} e\cdot$cm, (the current limit of reference \cite{Regan2002}), the energy scale is 26 mHz.

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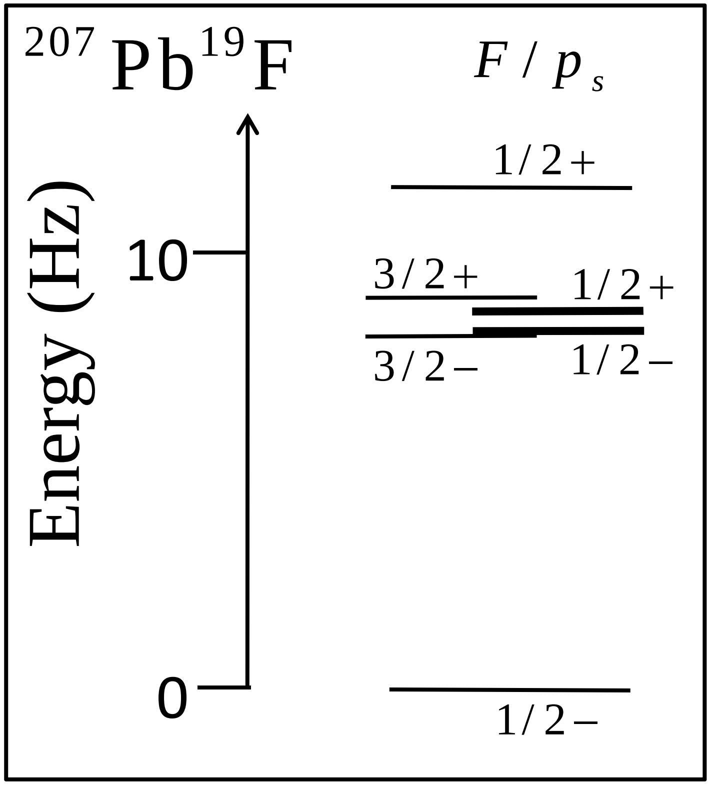
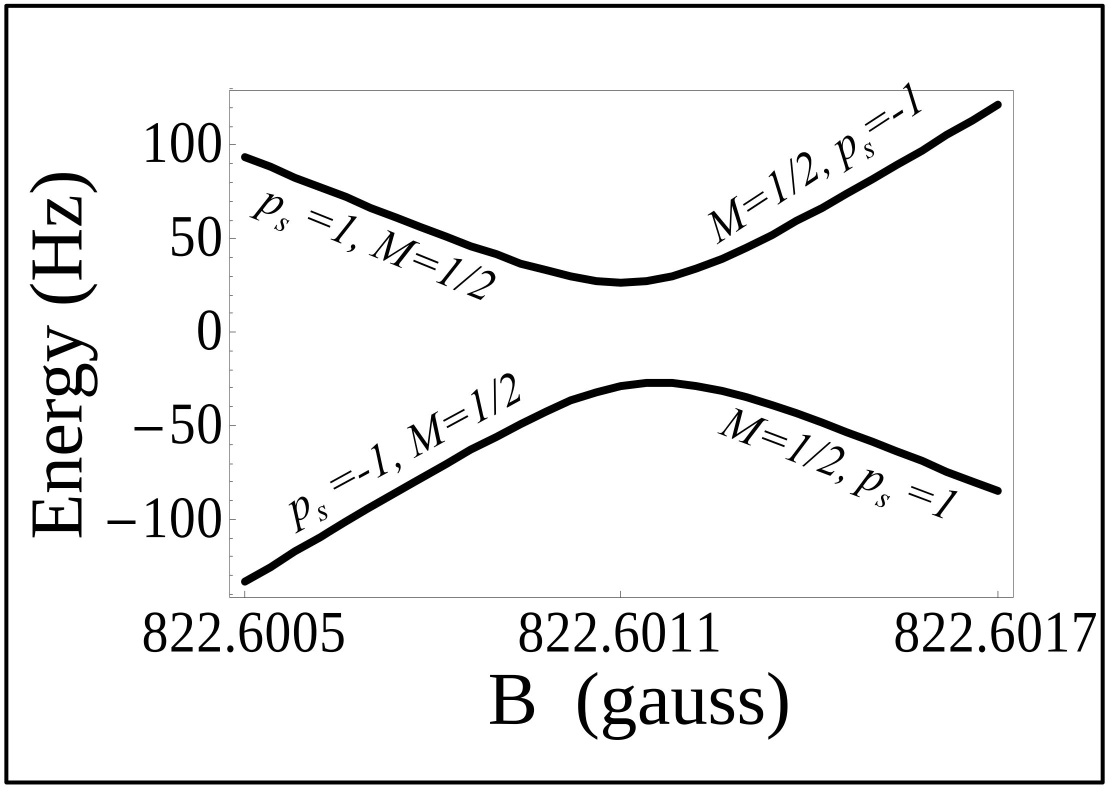
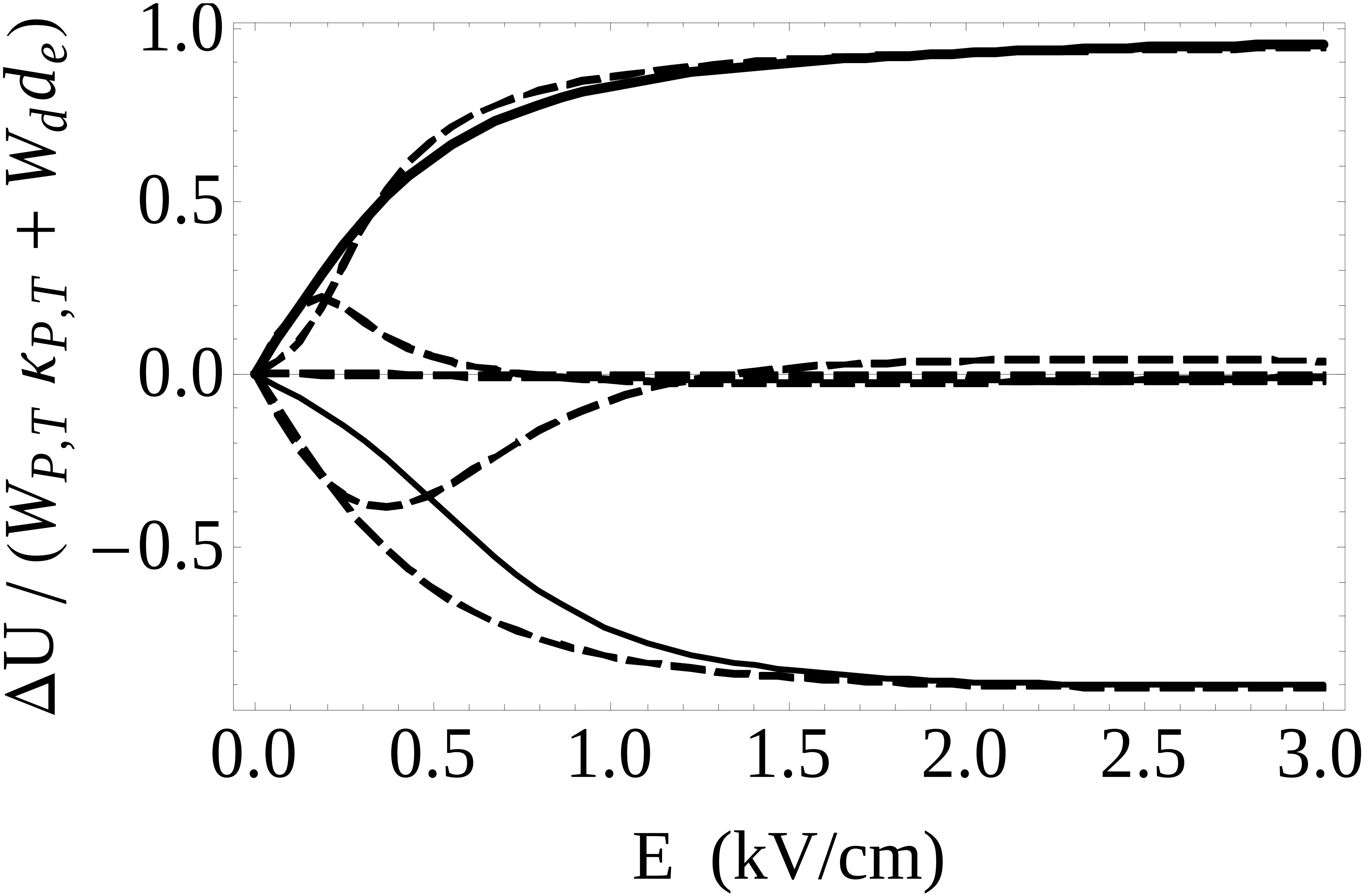


$^{207}Pb^{19}F$
$F\,/\,p_s$
Energy (Hz)
10
0
1/2+
3/2+
1/2+
3/2−
1/2−
1/2−



Energy (Hz)
100
50
0
−50
−100
822.6005
822.6011
822.6017
B (gauss)
p_s =1, M=1/2
M=1/2, p_s=-1
p_s=-1, M=1/2
M=1/2, p_s =1



ΔU / ($W_{P,T}\,\kappa_{P,T} + W_d d_e$)
1.0
0.5
0.0
−0.5
0.0
0.5
1.0
1.5
2.0
2.5
3.0
E (kV/cm)