Tenfold Improvement of Limits on T Violation in Thallium Fluoride

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We have made a stringent test of time-reversal symmetry using nuclear-spin resonance in a rotationally cold, supersonic beam of thallium fluoride molecules. We searched for a shift of the 120-kHz thallium spin resonance when a 29.5-kV/cm external electric field was reversed relative to the nuclear spin and found this to be $(1.4 \pm 2.4) \times 10^{-4}$ Hz. This is a tenfold improvement over our previous measurement in thallium fluoride. The derived constraints on the proton and electron electric dipole moments and on T violation in both strong and weak interactions are correspondingly improved.

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Although it is now 25 years since studies of kaon decay revealed that CP and T are not exact symmetries of nature[1] the underlying reason for the violation of these symmetries still remains obscure. In the hope of enlightenment, numerous searches have been made for any other manifestations of CP or T asymmetry. These include some very sensitive attempts to detect a permanent electric dipole moment (EDM) of a nondegenerate system, the existence of which would require the violation of both P and T symmetry. No EDM has been found, but the null results to data have nevertheless been important because of the strong restrictions they impose on theory.[2] At present the best limits on possible P- and T-asymmetric interactions outside the kaon system come from the neutron,[3] the Hg atom,[4] and the TlF molecule.[5] In this Letter we report an order-of-magnitude improvement in our experimental limits on T asymmetry in the thallium fluoride (TlF) molecule. This improvement leads to new constraints on several fundamental quantities.

Our experiment searches for a new kind of hyperfine interaction involving the spin-$\frac{1}{2}$ thallium nucleus (spin direction $\hat{\sigma}$) in TlF. The form of the effective Hamiltonian is

$$\mathcal{H}_{PT} = -hd\hat{\sigma}\cdot\hat{\lambda} \quad (1)$$

in which $\hat{\lambda}$ is a unit (polar) vector pointing from the Tl nucleus to the F nucleus, d is the coupling constant to be measured, and h is Planck's constant. This interaction is of interest because it violates both parity (P) and time-reversal (T) invariance and is of a rather general form.

Many different mechanisms are able, in principle, to induce an effective interaction of this kind.[6] One example is an EDM of the Tl nucleus, due either to an intrinsic nucleon EDM or to P-odd and T-odd nucleon-nucleon interactions. Another is a P-odd, T-odd electron-nucleon interaction. A third possibility is an intrinsic electric dipole moment of the electron. Our new measurement constrains the possible strengths of these effects.

The free TlF molecule is not directly sensitive to the interaction $\mathcal{H}_{PT}$ because the rotation of the molecule averages $\hat{\lambda}$ to zero. We therefore polarize $\hat{\lambda}$ along a 29.5-kV/cm external electric field **E**, which we can accurately reverse. A magnetic field **B**, parallel to **E**, defines an axis along which we polarize $\hat{\sigma}$ and this too can be independently reversed. Thus the signature of $\mathcal{H}_{PT}$ in our experiment is a shift of the energy levels that depends on the sign of $\hat{\sigma}$ and changes sign upon reversal of either **E** or **B** (but not both). In the experiment we look for a corresponding change Δf in the frequency of a NMR transition of the thallium nucleus.

The apparatus, shown schematically in Fig. 1, has been described in an earlier publication[5] and will be reviewed only briefly here. Molecules leaving the source in the rotational substate ($J=1$, $M_J=0$) are focused by an electrostatic quadrupole regardless of their nuclear spin state (M_{Tl} and M_{F}), while ($J=1$, $M_J=\pm 1$) molecules are defocused. The focused beam enters the first state selector where a combination of static and rf electric fields drives a transition to one of the $M_J=\pm 1$ sublevels. This transition is made in the presence of the magnetic **B**, which resolves all the substructure and allows us to populate a specific $(M_J, M_{\mathrm{Tl}}, M_{\mathrm{F}})$ sublevel of $J=1$, either $(-1,+\frac{1}{2},-\frac{1}{2})$ or $(+1,-\frac{1}{2},+\frac{1}{2})$. The main purpose of this state selector is to polarize $\hat{\sigma}$, i.e., to select M_{Tl}.

The NMR transition of the thallium nucleus is then induced using separated oscillating magnetic fields in the presence of the strong electric field **E**. A second state selector and quadrupole render the NMR transition observable by focusing onto a hot-wire detector those molecules that did not undergo a thallium spin flip and by de-

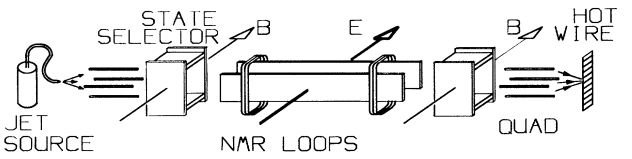

FIG. 1. Schematic view of the apparatus.

focusing those that did. The NMR signal is observed as a change in the detector current when the phase angle ϕ between the separated oscillating fields is switched from $+\pi/2$ to $-\pi/2$. Figure 2 shows the NMR signal that we measure as the oscillator frequency is swept through the resonance.

The tenfold improvement reported here was made possible by two main experimental advances. First, we developed a new design of supersonic jet source; we are now able, without the use of any carrier gas, to obtain an intense beam of rotationally cold TlF monomers. This source increased the strength of our NMR signal by a factor of 100 while (transit time) broadening the line by only 50%. Furthermore, the noise measured in the beam is now only slightly above the shot-noise limit. The signal-to-noise ratio of our experiment has so far increased by a factor of 14 as a result of the new source. The second important development was the use of an additional reversal which enhanced our ability to discriminate against systematic errors and which we now discuss.

Four reversals are employed to identify Δf, the contribution to the hyperfine interval arising from $\mathcal{H}_{PT}$. (i) Effects associated with the nuclear resonance are isolated by switching the relative phase ϕ of the separated NMR fields. (ii) $\hat{\boldsymbol{\lambda}}$ is reversed by switching **E**. (iii) All the angular momenta in the molecule, including $\hat{\boldsymbol{\sigma}}$, are reversed by inverting the quantization axis **B** in the state selectors. (iv) The angular momenta are also reversed by changing the frequency of the rf field in the state selectors. Previously the selected state was always $(M_J, M_{\mathrm{Tl}}, M_{\mathrm{F}}) = (-1, +\frac{1}{2}, -\frac{1}{2})$, whereas now we can switch to $(+1, -\frac{1}{2}, +\frac{1}{2})$—the "time-reversed" state—as well, without changing any of the static fields. This last reversal is the new addition to our experimental method. We call it the "**M**" reversal because it is a reversal of the magnetization of the molecule.

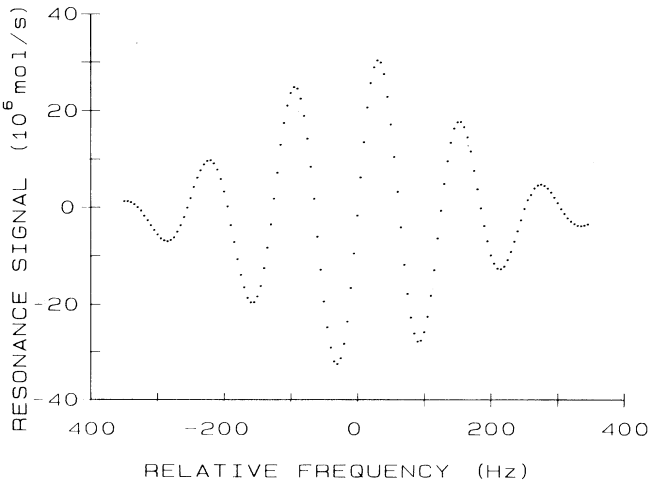

FIG. 2. Observed NMR signal vs frequency. Ordinate is the change at the detector (in units of 10^6 molecules per second) when the phase between the separated oscillating fields is switched from $\pi/2$ to $-\pi/2$.

The measurement of $\mathcal{H}_{PT}$ was conducted close to the central zero crossing of Fig. 2, where the NMR signal is least sensitive to a change in the beam intensity and most sensitive to genuine shifts of the resonance frequency. The four independent reversals (ϕ, **B**, **E**, and **M**) that were used to isolate Δf generated a total of sixteen possible configurations of the apparatus. We spent half the time at a frequency 1.25 Hz above the central zero crossing and the other half equally below in order to measure the slope of the resonance, and at each frequency we devoted equal time to all sixteen configurations.

The modulations were controlled by a computer operating in a series of nested loops $E\{B\{F\{M\{\phi\}\}\}\}$. The innermost loop switched the relative phase ϕ of the NMR fields in a pattern $(+--+-++-)$ of eight, 50-ms "data intervals" and recorded the integrated detector current for each data interval. This pattern was chosen to discriminate against linear and quadratic drifts of the beam intensity. The next loop controlled the **M** reversal according to the pattern $(+--+)$, while the third loop set the oscillator frequency (F) to be above or below the zero crossing also in the sequence $(+--+)$. The fourth loop controlled the magnetic field **B** and the outermost loop switched the electric field **E** according to the sequence $(+--+)$. Completion of the outermost loop constituted a "measurement" of Δf. The pattern for switching **B** alternated between $(+--+)$ and $(-++-)$ for successive measurements. Thus each measurement consisted of 2048 data intervals and represented 102.4 sec of data acquisition. The real time devoted to each measurement was typically 240 sec because some additional time was needed to gate out beam transients associated with the reversals.

A typical run consisted of twenty measurements, taken one after the other under identical conditions over a period of some 90 min. The P-odd, T-odd frequency shift Δf was determined from that part of the detector current which varied synchronously with the switching of ϕ and with the reversals of **E**, **M**, and **B**. Our determination of Δf was based on a set of 560 such measurements taken in a series of 28 runs. The distribution of these measurements within a single run was well described by a single Gaussian whose width was on average only 14% larger than the shot-noise limit. This width varied from one run to another because of changes in the beam intensity when the apparatus was realigned and detector efficiency when the hot wire was replaced. The weighted mean of the 28 runs and the standard deviation of the mean are

$$\Delta f = +0.14 \pm 0.24 \text{ mHz} \tag{2}$$

and this result is illustrated by the shaded area of Fig. 3. It is clearly consistent with zero.

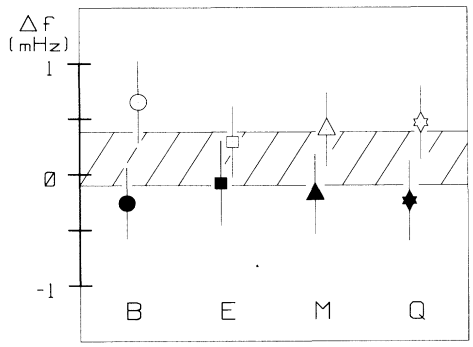

FIG. 3. Summary of measurements. Shaded area indicates the final result. Data points show results grouped according to the manual reversals **B**, **E**, **M**, and **Q** (defined in the text) and indicate the absence of systematic effects.

Between each run and the next, we made a manual reversal of either **E**, **B**, or **M**. This involved interchanging the wires from the high-voltage supplies, magnetic field supplies, or state-selector oscillators. These supplementary reversals allowed us to check for systematic effects related to the state of the switches or the data-collection electronics rather than to the state of the fields in the apparatus. We also looked for a systematic effect associated with an interchange of the negative and positive connections to the quadrupole lenses (**Q**). Figure 3 shows the mean and standard deviations obtained when our measurements are split into two sets according to the manual reversals of **B**, **E**, **M**, and **Q**. We conclude that there are no significant systematic shifts of this type.

Another potential source of error is the inexactitude of the automated reversals **E**, **M**, and **B**. For example, when we try exactly to reverse the electric field in the apparatus, its magnitude is altered by a small amount, both in the state selectors and in the NMR region. This changes both the strength and the frequency of the NMR signal in synchronism with the E reversal. Similar effects are associated with imperfections of the **M** and **B** reversals. Fortunately, the shift Δf is insensitive to these imperfections in the first approximation. Even so, we have worked to make them small; the amplitude changes are all less than 1% and the frequency shifts are all less than 10 mHz. At a smaller level (second order), the amplitude change associated with one reversal can conspire with the frequency shift of another to produce a possible false Δf. Although this effect can be measured and corrected (the correction was 0.7 mHz in our first experiment[5]) a third reversal provides a more satisfactory remedy, for now the false shift cannot contribute to the result except at the level of third order in the imperfections. In this work the correction to our final result was only -0.06 mHz, which is much less than the random error. Furthermore, we believe it can be significantly reduced for future measurements by shielding the NMR region more carefully from the magnetic field of the state selectors.

The measured frequency shift is simply related to the P-odd, T-odd coupling constant d:

$$\Delta f = -2d\,|\langle\hat{\boldsymbol{\lambda}}\rangle|\,. \tag{3}$$

In the 29.5-kV/cm field of this experiment $|\langle\hat{\boldsymbol{\lambda}}\rangle| = 0.542$. Hence the result of our experiment is

$$d = -0.13 \pm 0.22\ \text{mHz}\,. \tag{4}$$

Now we turn to the possible interpretations of Eq. (4) in terms of more fundamental quantities. First, our value for d can be used to place a limit on any interactions that generate an electric dipole distortion of the Tl nucleus. Although the level shift due to this dipole is suppressed in accordance with Schiff's theorem,[7] there remains in the atomic Hamiltonian an interaction of the form[7-9]

$$\mathcal{H}_{PT} = -4\pi e\nabla\rho(0)\cdot\mathbf{Q} \tag{5}$$

in which $\nabla\rho(0)$ is the gradient of electron density at the nucleus and lies along $\hat{\boldsymbol{\lambda}}$, and the Schiff moment $\mathbf{Q}$ is related to the nuclear dipole moment and lies along $\boldsymbol{\sigma}$. Using the theoretical value[8] $\langle 4\pi\nabla\rho(0)\rangle = 1.63\times10^{4}a_0^{-4}\hat{\boldsymbol{\lambda}}$, we find that the Schiff moment of Tl is

$$Q_{\mathrm{Tl}} = (-1.8 \pm 3.0)\times10^{-23}\,e\,\text{cm}\,\text{fm}^2\,. \tag{6}$$

This result constrains the strength of T-odd, P-odd, nucleon-nucleon interactions[10] to the level of 10^{-2} of the Fermi constant G_F. Similar limits are implied by the measurements on the Hg atom and on the neutron.[11] In addition, we are able[8,12] to place a much improved limit on the electric dipole moment of the proton:

$$d_p = (-3.7 \pm 6.3)\times10^{-23}\,e\,\text{cm}\,. \tag{7}$$

In a second type of interpretation, we can place limits on the coupling strength of possible semileptonic interactions such as weak electron-nucleon point interactions of the form[13]

$$\mathcal{H}_{PT} = iC_T\frac{G_F}{\sqrt{2}}(\bar{n}\sigma^{\mu\nu}n)(\bar{e}\gamma_5\sigma_{\mu\nu}e)\,, \tag{8a}$$

$$\mathcal{H}_{PT} = iC_S\frac{G_F}{\sqrt{2}}(\bar{n}n)(\bar{e}\gamma_5 e)\,, \tag{8b}$$

where n and e are the nucleon and electron field operators. The possibility of placing a limit on C_S may seem surprising at first sight since the interaction involves an axial electron current and yet the electron spin does not appear to have any role in our experiment—in fact, the electronic shells of the TlF molecule are closed ($^1\Sigma_0$). However, Flambaum and Khriplovich[14] have pointed out that an axial electron current can lead to an effective nuclear dipole [i.e., $d\neq0$ in Eq. (1)], even in a system of closed electron shells, as a result of the hyperfine interaction.

Following Refs. 8 and 14, and assuming for simplicity

that the interactions are isoscalar, we find

$$C_T = (1.7 \pm 3.0) \times 10^{-7}, \tag{9a}$$

$$C_S = (5.4 \pm 9.2) \times 10^{-6}. \tag{9b}$$

In the purely leptonic sector, we are able to restrict the electric dipole moment of the electron,[14] again through the hyperfine coupling to the nuclear spin. The result is

$$d_e = (-1.4 \pm 2.4) \times 10^{-25}\, e\,\text{cm}. \tag{10}$$

Our results for C_T and C_S are approximately equal to the best previously available.[4] The value of d_e is 5 times less restrictive than the very recent result obtained by Murthy *et al.*[15] but 5 times smaller than the best previous limit.[6] Our limits on Q_{Tl} and d_p are an order of magnitude smaller than any other.

These first results using a supersonic beam of TlF have not revealed a new case of T asymmetry but have substantially improved the experimental limits. Moreover, the full potential of our method has still to be developed and we can expect at least another factor of 10 reduction in the uncertainty over the next few years.

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