Sensitivity of condensed-matter *P*- and *T*-violation experiments

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Experiments searching for parity- and time-reversal-invariance-violating effects that rely on measuring the magnetization of a condensed-matter sample induced by application of an electric field are considered. A limit on statistical sensitivity arises due to random fluctuations of the spins in the sample. The scaling of this limit with the number of spins and their relaxation time is derived. Application to an experiment searching for nuclear Schiff moment in a ferroelectric is discussed.

I. INTRODUCTION

Much of the present knowledge about the fundamental symmetries *CP* (invariance under combined operations of spatial inversion and charge reversal) and *T* (invariance with respect to time reversal) comes from experiments measuring *P*- and *T*-violating permanent electric-dipole moments (EDM's) of atoms, molecules, and the neutron; see, for example, Ref. [1]. Most EDM experiments measure precession of the angular momentum of the system in an applied electric field analogous to the Larmor precession in an applied magnetic field.

In addition to such *precession experiments*, there are EDM searches of another kind [2,3], which have drawn recent renewed attention [4–7]. The idea of these experiments is the following. Suppose that we have some condensed-matter sample that has $\mathcal{N}$ spins (either electron or nuclear depending on the specific experiment). If an electric field is applied to the sample, it interacts with the associated (*P*- and *T*-violating) EDM's, leading to a slight orientation of the spins in the direction of the electric field. This orientation, in turn, is measured by measuring the induced magnetization of the sample.

In this paper, we compare the precession and condensed-matter experiments in the high- and low-temperature limits (Secs. II and III), discuss the magnetic-field noise in light of the fluctuation-dissipation theorem (FDT) (Sec. IV), discuss the implications of the present analysis for a proposed search for the nuclear Schiff moment in a ferroelectric material (Secs. V and VI), and summarize our conclusions in Sec. VII.

II. COMPARISON OF THE PRECESSION EDM EXPERIMENTS WITH CONDENSED-MATTER EXPERIMENTS IN THE HIGH-TEMPERATURE LIMIT

The signal in "traditional" EDM experiments is given by

$$S_1 \approx \mathcal{N}\frac{dE}{\hbar}\tau. \tag{1}$$

Here $\mathcal{N}$ is the number of particles involved in a measurement, *d* is the EDM, *E* is the effective electric field acting on the particle, and τ is the spin-relaxation time. This assumes a "single-shot Ramsey-type" measurement scheme where the particles are polarized, precess in the electric field, and then their precession is probed with high efficiency after a time on the order of the spin-relaxation time. The noise of such a one-shot measurement is

$$N_1 \approx \sqrt{\mathcal{N}}. \tag{2}$$

The corresponding *S*/*N* ratio can be improved by repeating the measurements many times up to a total experiment time *t*:

$$S/N \approx \frac{S_1}{N_1}\sqrt{\frac{t}{\tau}} = \sqrt{\mathcal{N}}\frac{dE}{\hbar}\sqrt{\tau t}. \tag{3}$$

Let us now consider condensed-matter experiments measuring magnetization induced by application of an electric field. Let us say, for the sake of the argument, that we have an ideal noise-free external magnetometer with unlimited sensitivity. What is the statistical sensitivity of the experiment?

The EDM-induced magnetic moment of the sample is given by

$$S_1 \propto \mathcal{N}\frac{dE}{T}\mu, \tag{4}$$

where *E* is the effective electric field acting on the spins, *T* is the absolute temperature of the spins in energy units, and μ is the magnetic moment of one spin. This is our signal. What is the noise?

In the absence of any external fields, at a given moment in time we have a random total magnetic moment

$$N_1 \propto \sqrt{\mathcal{N}}\mu. \tag{5}$$

As in the case of a precession experiment, the fact that this noise magnetic moment is random and changes in time may

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be used to improve the S/N ratio. In order to characterize the correlation time of the fluctuations, we introduce spin-relaxation time τ. This parameter characterizes how long the random magnetic moment persists. If the time τ is long, this may present a serious problem for the experiment. In other words, if the spins relax slowly, there is a random signal, which would not average to near zero in a short time.

More formally, we have expressions (4) and (5) from which we can write S/N for a long measurement time $t \gg \tau$:

$$S/N \approx \frac{S_1}{N_1}\sqrt{\frac{t}{\tau}} = \sqrt{\mathcal{N}}\frac{dE}{T}\sqrt{t/\tau}. \tag{6}$$

This shows that the key parameters for an experiment of this type are the relaxation time τ and the temperature. Assuming that these parameters are independent, the experiment should be done at the lowest possible temperature to increase the degree of induced polarization. In addition, it appears that it may be beneficial to have *fast* spin relaxation (small τ), so that the measurement can be repeated often. Such dependence of the sensitivity on τ is the opposite of that in the case of precession experiments [Eq. (3)].

III. WHAT HAPPENS AT LOW TEMPERATURE: THE USUAL SCALING RECOVERED

Let us now consider a case where relaxation is determined by the interaction between the spins—the dipole-dipole interaction (see Sec. V). The characteristic energy scale $\mathfrak{J}$ for such an interaction is related to the relaxation time according to

$$\mathfrak{J} \approx \frac{\hbar}{\tau}. \tag{7}$$

It is now important to mention that in the presence of such a residual interaction, our assumption that the induced magnetization is inversely proportional to the temperature breaks down when the temperature becomes comparable to the residual interaction. Depending on the details of the interactions, the spin system can go into, for example, a ferromagnetic or antiferromagnetic state for $T < \mathfrak{J}$ (see, for example, Ref. [8]), upon which the susceptibility vanishes and the system is no longer sensitive to EDM's. This effect limits the optimal temperature of the sample to

$$T_{opt} \approx \mathfrak{J}. \tag{8}$$

Substituting this into Eq. (6) and taking into account Eq. (7), we recover a result that is identical to that of Eq. (3) for "traditional" precession EDM experiments.

IV. MAGNETIC-FIELD NOISE AND THE FLUCTUATION-DISSIPATION THEOREM

The energy associated with the spins in a polarized paramagnetic material (this could be nuclear paramagnetism as in the case of the Schiff-moment experiment proposed in Ref. [6]) can be written as

$$\mathcal{E} = -\frac{1}{2}\mathbf{M} \cdot \mathbf{B}V, \tag{9}$$

where $\mathbf{B}$ is the magnetic induction (assumed uniform in a volume V) and $\mathbf{M} = \chi\mathbf{B}$ is the average induced magnetization; χ is the paramagnetic susceptibility. This direct linear link between M and B suggests that the fluctuations can be determined from the FDT after we have ascertained a generalized susceptibility (see Ref. [9], Sec. 124).

The spins become polarized after application of a magnetic field due to a dissipative process; the spins relax to the equilibrium polarization through the *longitudinal* relaxation characterized by a time constant T_1.

Let us discuss the steady-state response of the magnetization to an oscillatory magnetic field applied to the sample at a frequency ω (with no applied static field). The specific form of the response depends on the system. We consider two models representing a range of possible high-frequency behaviors of the susceptibility:

$$\text{(i)} \quad \text{Im}[\chi(\omega)] = \chi_0 \frac{\omega T_1}{1 + \omega^2 T_1^2}, \tag{10}$$

$$\text{(ii)} \quad \text{Im}[\chi(\omega)] = \chi_0 \sqrt{\pi}\omega T_1 e^{-\omega^2 T_1^2}. \tag{11}$$

Here χ_0 is the usual Curie susceptibility

$$\chi_0 \approx \frac{\rho\mu^2}{T}, \tag{12}$$

where the angular momentum factors have been neglected, ρ is the number density, and the temperature T is expressed in energy units. The full complex susceptibility can be reconstructed using the Kramers-Kronig relations (see, for example, Ref. [9], Sec. 123):

$$\chi(\omega) = -\frac{1}{\pi}\int_{-\infty}^{+\infty}\frac{\text{Im}[\chi(x)]}{\omega - x + i0}dx. \tag{13}$$

This gives

$$\text{(i)} \quad \chi(\omega) = \chi_0 \frac{1}{1 - i\omega T_1}, \tag{14}$$

$$\text{(ii)} \quad \chi(\omega) = \chi_0 \begin{cases} 1 + i\sqrt{\pi}\omega T_1 - 2(\omega T_1)^2 + \cdots, & \omega T_1 \ll 1, \\ -1/(2\omega^2 T_1^2) + \cdots, & \omega T_1 \gg 1. \end{cases} \tag{15}$$

The first model is more relevant to electron spins and to nuclear spins when they are coupled to the lattice (see also Refs. [10,11] for treatments of similar problems). The second model is more relevant to nuclear spins in an insulator at a sufficiently low temperature when the spins are decoupled from the lattice [12]. We are interested in the low-frequency regime $\omega T_1 \ll 1$. In this regime both models give the same results. To be specific below we use the first model. (This form of the response is analogous to the behavior of an electrical RC circuit. In the low-frequency limit $\omega T_1 \ll 1$, the susceptibility tends to its static limit χ_0 and the magnetization is in phase with the induction. In the high-frequency limit,

$\omega T_1 \gg 1$, magnetization is $\pi/2$ out of phase with induction, with its magnitude scaling inversely proportionally with the frequency.)

We are now poised to directly apply the FDT to this problem and write an expression for the spectral density $(M^2)_\omega$ of the square of the deviation of the magnetization from its average value:

$$V(M^2)_\omega = \hbar \ \coth(\hbar\omega/2T)\text{Im}\chi(\omega) \approx \frac{2\chi_0 T_1 T}{1 + (\omega T_1)^2}. \quad (16)$$

In the last part of the above expression, we have used $\coth(\hbar\omega/2T) \approx 2T/\hbar\omega$, which is true for $T \gg \hbar\omega$. Combining the final expression of Eqs. (16) and (12), we get

$$V(M^2)_\omega \approx \rho\mu^2 \frac{2T_1}{1 + (\omega T_1)^2}. \quad (17)$$

There are several properties of this expression that should be noted. First, the average square magnetization is inversely proportional to the volume of the sample. This represents averaging of fluctuations over parts of a large sample. Another remarkable result is that the magnetization noise has no temperature dependence other than through a possible temperature dependence of T_1. This is because the noise energy goes as T but the susceptibility goes as $1/T$, so the two factors cancel.

For a properly optimized geometry of a solid-state EDM experiment, the detected signal depends on the magnetic moment of the entire sample. For an experiment with an averaging time $t \gg T_1$, the ongoing analysis reproduces the scaling of Eq. (6) if we identify the relevant relaxation time τ with T_1. (It is the transverse relaxation T_2 that is of relevance in precession experiments.) Indeed, estimating

$$(M^2)_\omega \approx M^2 T_1, \quad (18)$$

setting $\omega = 1/T_1$, multiplying both sides of Eq. (17) by V, and taking the square root, we reproduce the noise of Eq. (5).

V. SOME FEATURES OF THE PROPOSED NUCLEAR SCHIFF-MOMENT EXPERIMENT

In this section we discuss some peculiar features of nuclear Schiff-moment experiments in ferroelectric solids proposed in Ref. [6].

We consider a diamagnetic solid-state system with nonzero-spin nuclei (these are the nuclear-spin $I = 1/2$ ^{207}Pb nuclei with magnetic moment of $0.59\mu_N$, where μ_N is the nuclear magneton, in the specific proposal involving ferroelectric lead titanate). The lattice temperature is always considered low enough, so the effect of phonons and, specifically, the interaction between the nuclear spins and the lattice mediated by lattice vibrations are completely negligible. In practical terms, this would require cooling the sample to temperatures on the order of a kelvin.

Under such conditions, the lattice is decoupled from the nuclear spins with the exception of the fact that the spins are "pinned" to the lattice. Assuming that the spins only interact with each other (by means of sensing each other's magnetic field) and that there is no interaction with the lattice other

than that the lattice keeps the nuclei fixed in space, it is straightforward to estimate the spin-relaxation rate (see, for example, Ref. [13], Chap. 13). Because the magnetic field from a dipole falls as the inverse third power of the distance, for a given spin, relaxation is determined by its closest neighbor(s). The relaxation rate can be estimated as the Larmor precession rate of a spin in its neighbor's field:

$$\gamma \sim \frac{(\mu_N)^2}{\hbar r^3}. \quad (19)$$

Here μ_N is the nuclear magneton and r is the characteristic distance between the neighbors. If the distance between interacting spins is on the order of interatomic spacing in condensed matter, the relaxation rate is on the order of kilohertz. This relaxation provides a lower limit on the magnetic-resonance linewidth. For the specific case of lead titanate, the dipole-dipole relaxation rate is estimated in Ref. [6] as being $\gamma/(2\pi) \approx 200$ Hz. It is important that despite the fact that the nuclear spins are isolated from the lattice, the total angular momentum of the nuclear spin-system is not conserved. This is easy to see from the following argument involving, for simplicity, just two spins.

The Hamiltonian describing the interaction between the spins is

$$\hat{H} = -\vec{\mu}_1 \cdot \vec{B}_{21} = -\vec{\mu}_1 \cdot \frac{3(\vec{\mu}_2 \hat{r}_{12})\hat{r}_{12} - \vec{\mu}_2}{r_{12}^3}. \quad (20)$$

Here $\vec{\mu}_{1,2} = g_{1,2}\mu_N \vec{I}_{1,2}$ are the magnetic moments of the two spins, $g_{1,2}$ are their nuclear g factors, $\vec{I}_{1,2}$ are their spin operators, $\vec{r}_{12}$ is the separation between the spins, and $\hat{r}_{12}$ is the unit vector in the direction of $\vec{r}_{12}$.

Let us examine whether the total spin projection $M_1 + M_2$ onto a given quantization axis is a conserved quantity. To do this, we check whether the corresponding operator $I_z = I_{1z} + I_{2z}$ commutes with the Hamiltonian of Eq. (20):

$$[I_z, \hat{H}] = \frac{-g_1 g_2 \mu_N^2}{r_{12}^3}[I_{1z} + I_{2z}, 3(\vec{I}_1 \cdot \hat{r}_{12})(\vec{I}_2 \cdot \hat{r}_{12}) - \vec{I}_1 \cdot \vec{I}_2].$$

$$(21)$$

The commutator term $[I_{1z} + I_{2z}, \vec{I}_1 \cdot \vec{I}_2]$ is zero, but the other term in Eq. (21) is generally not. This is because, for example, $\vec{I}_1 \cdot \hat{r}_{12}$ is a linear combination of the operators I_{1x}, I_{1y}, and I_{1z}, the first two of which do not commute with I_{1z}.

Thus we see that the total spin angular momentum is not conserved in dipole-dipole interactions and the angular momentum is exchanged with the lattice. A detailed discussion of the evolution of systems of many spins on a lattice has been given in Ref. [14].

The scale of the dipole-dipole interaction strength $\mathfrak{J}$ expressed in temperature units corresponds to tens to hundreds of nanokelvins. As discussed in Sec. III, the EDM experiment would ideally be conducted at spin temperatures slightly higher than this.

At this point, prior to proceeding with the discussion of the EDM measurement, let us consider several thought experiments that will help in understanding of the spin system.

We first assume that a strong magnetic field is initially applied, so the spins are polarized. (It is not necessary that the field be strong enough to lead to full polarization, but it has to be much stronger than the characteristic value of the dipole field.) We then turn off the leading field abruptly. The question is to which state does the system relax and at what rate?

The way we have set up the problem, the magnetic interaction between the spins is the only interaction affecting the spins, so the spin polarization will relax at a rate on the order of $\mathfrak{J}$ (where we do not distinguish between energy, temperature, and frequency units). Because, as discussed above, angular momentum is not conserved, the final state of the spins will have no average polarization. The temperature of the spins will remain the same. In this state, each of the spins "sees" a randomly fluctuating field from other spins, which has a characteristic correlation time of $1/\mathfrak{J}$ and just the appropriate characteristic magnitude that it rotates the spin (via Larmor precession) by an angle of order unity during a correlation time. Consequently, the overall magnetic moment randomly oscillates with the same correlation time and the overall magnitude proportional to the square root of the total number of spins in the sample as discussed in the preceding sections. These time-dependent fluctuations are essential for the Schiff-moment experiment as they serve to average the random polarization of the sample, while preserving the "bias" due to the *P*- and *T*-violating effect.

An interesting question is, what happens if the strong magnetic field is reapplied quickly (much faster that the correlation time)? After the field is turned on, the magnitude of this strong field is much greater than the dipole fields and each of the spins precesses around the direction of the strong field. Effectively, in this regime, the components of the dipole fields perpendicular to the leading field have no effect on the spins and the only effect of the longitudinal components is to produce a small variation of the overall field magnitude from site to site. Such inhomogeneous broadening is important for transverse (T_2) relaxation, but is irrelevant for longitudinal (T_1) relaxation. Thus, after application of the strong field, the spin system remains in the unpolarized state[1] indefinitely in the framework of the approximations that we have assumed here. In practice, some slow T_1-relaxation processes will eventually relax the spins into a state where their magnetic moments are preferentially along the strong leading field, which is the equilibrium state. Note that such behavior of the nuclear-spin subsystem isolated from the lattice was discussed already half a century ago in Ref. [15].

Next, we discuss how the nuclear spin-system can be cooled to a low temperature (the desired temperature is slightly above $\mathfrak{J}$). This will require a slow decrease of a leading field as opposed to rapid leading-field variations. Suppose an experimentally realizable magnetic field of $B = 10^5$ G is applied and the sample is cooled down to a temperature $T_0 \sim 1$ K where the nuclear spins decouple from the

lattice. At this point, the polarization for the case of ^{207}Pb is

$$\approx \frac{\mu B}{T_0} \sim 10^{-3}. \tag{22}$$

The magnetic field is then slowly turned off, causing adiabatic-demagnetization cooling of the nuclear spin system. The spin temperature at the end of cooling can be estimated as

$$T \sim T_0 \frac{J}{\mu B} = T_0 \left(\frac{\mu B}{T_0} \right)^{-1} \frac{\mathfrak{J}}{T_0} \sim 10^{-5} - 10^{-4} \text{ K}. \tag{23}$$

Unfortunately, due the smallness of the factor (22), this is a significantly higher temperature than the desired $\gtrsim \mathfrak{J}$.

VI. ESTIMATE OF THE STATISTICAL SENSITIVITY OF THE SCHIFF-MOMENT EXPERIMENT

Let us take the nuclear spin temperature $T = 10^{-4}$ K, a conservative estimate in Eq. (23). The magnetic moment of a ferroelectric lead-titanate sample induced by a Schiff moment, according to Eq. (8) of Ref. [6], is

$$VM \approx 10^6 \mathcal{N} \mu S \frac{1 \text{ eV}}{T} \sim 10^{14} \mathcal{N} \mu S. \tag{24}$$

Here the Schiff moment S of the ^{207}Pb nucleus should be expressed in units of $e \, a_0^3$.

Estimating the signal-to-noise ratio (assuming noise-free magnetometer) along the lines of the discussion in Sec. II, we have

$$S/N \sim 10^{14} \sqrt{\mathcal{N}} S \sqrt{\gamma t} \sim 10^{30} S. \tag{25}$$

For the final step of the estimate (25), we have taken $\mathcal{N} = 3.3 \times 10^{22}$ corresponding to a volume of $V = 10$ cm^3 and the 22.1% natural abundance of ^{207}Pb; the experiment duration of $t = 10$ days and $\gamma \approx 10^{-12}$ eV [in frequency units, $\gamma/(2\pi) \approx 200$ Hz]. Thus, an $S/N = 1$ corresponds to a sensitivity to the Schiff moment of approximately $10^{-30} e \, a_0^3$. This is by more than four orders of magnitude better than the present best limits on the Schiff moment of ^{199}Hg (see Ref. [16] and references therein).

Finally, it is interesting to estimate a characteristic magnitude of the spin-noise magnetic field. Assuming a sample with all characteristic dimensions $2R$, just outside of it, the noise magnetic field is on the order of

$$B_N \sim \frac{\sqrt{\mathcal{N}} \mu}{R^3} \sim 10^{-12} \text{ G}. \tag{26}$$

The noise produced by the spins is comparable to the noise of modern magnetometers; see, for example, Ref. [17] and references therein.

VII. CONCLUSION

In this paper, we have considered EDM experiments that rely on measuring the magnetization of a condensed-matter sample induced by application of an electric field. A limit on the statistical sensitivity of such an experiment arises due to

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[1]More precisely, the system remains in a state with a random weak polarization along the leading field equal to the polarization component in this direction that existed due to fluctuations at the moment when the strong field was turned on.

random fluctuations of the spins in the sample. We find that, while the ultimate sensitivity has the usual scaling ($\propto \sqrt{\mathcal{N}t}$) with the number of spins and the measurement time, in the limit where the temperature greatly exceeds the spin-spin interaction energy, the sensitivity also scales $\propto \sqrt{\tau/T}$. Such scaling with relaxation time is radically different from that for the more traditional precession EDM experiments. Interestingly, the usual scaling is recovered if one is able to cool the spins to a low temperature, comparable to the energy of the magnetic dipole-dipole interaction between the spins.

After presenting a heuristic derivation of this result, we have discussed how it can be obtained from the fluctuation-dissipation theorem.

Finally, we have presented an estimate based on the earlier results of Ref. [6] combined with the present considerations of the noise due to spin fluctuations of the ultimate statistical sensitivity of a search for the *P*- and *T*-odd nuclear Schiff moment using a ferroelectric material. We find that, with realistic experimental parameters, the statistical noise due to spin fluctuations should not preclude obtaining a significant improvement in sensitivity to the Schiff moment

(perhaps, up to four orders of magnitude) compared with the present best limits.

An important limiting factor for the nuclear Schiff-moment experiment appears to be the difficulty of cooling the spin system to a sufficiently low temperature (in the tens of nanokelvin range) using the adiabatic demagnetization technique. The limitation comes from the fact that thermal polarization of the spins in an achievable laboratory magnetic field is very low. In principle, it may be possible to produce much higher initial nuclear-spin polarizations, for example, by creating UV-light-induced metastable paramagnetic centers [18] and performing optical pumping.

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