Macroscopic T Nonconservation: Prospects for a New Experiment

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Breakdown of time-reversal invariance can be detected in macroscopic samples as a magnetic alignment along an electric field. We show that both fundamental and practical limits to the detection of this effect in paramagnets correspond to measurement of electron electric dipole moments down to $d_e \sim 10^{-28}$ $e \cdot$cm on 50-g quantities of EuS near its Curie point; this compares to the current limit of $d_e \leq 10^{-24}$ $e \cdot$cm. Strategies for still greater sensitivity are outlined.

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Evidence for the existence of interactions which violate[1] CP and[2] T symmetries has been found thus far only in the celebrated K-$\bar{K}$ system. These experiments alone, however, do not provide the constraints needed to discriminate among the various models which have been proposed to explain CP and T nonconservation.

One consequence of T and P nonconservation is the possible existence of electric dipole moments (EDM's) for the elementary particles.[3] As yet, however, no positive evidence for the existence of EDM's has been found. Current EDM limits for the neutron and electron are $d_n < 10^{-24}$ $e \cdot$cm from neutron-beam experiments,[4] $d_n < 6\times 10^{-25}$ $e \cdot$cm from experiments with bottled neutrons,[4] and $d_e < 10^{-24}$ $e \cdot$cm from gas-phase atomic spectroscopy and atomic-beam experiments.[5] These limits should be compared with typical theoretical predictions. For the neutron, experimentally allowed predictions range from $d_n \sim 10^{-24}$ $e \cdot$cm for Higgs-sector models to $d_n \sim 10^{-28}$ $e \cdot$cm for models with additional quarks,[4] down to $d_n \sim 2\times 10^{-32}$ $e \cdot$cm for the Kobayashi-Maskawa model.[6] For the electron, models based upon scalar-lepton couplings can yield EDM's as high as $d_e \sim 4\times 10^{-25}$ $e \cdot$cm,[7] models based on massive Dirac neutrinos or family symmetries predict upper bounds $d_e < 10^{-27}$ $e \cdot$cm,[8] and Higgs-sector models yield the lowest finite estimates of $d_e \sim 10^{-32}$ $e \cdot$cm.[4]

Current bounds on the EDM's of elementary particles have resulted largely from the study of "one particle at a time." It is reasonable to ask whether sensitivity can be enhanced by the study of macroscopic numbers of particles. The macroscopic approach was first suggested by Fairbank,[9] who proposed a search for nuclear EDM's in dilute solutions of ^{3}He in liquid ^{4}He. Ignatovich[10] extended these ideas by suggesting searches for d_e in ferromagnets; a subsequent experiment by Vasilev and Kolycheva[11] resulted in $d_e < 10^{-22}$ $e \cdot$cm. Other proposals for macroscopic experiments include searches for P-nonconserving e-N neutral-current interactions in ^{3}He-B,[12] for nuclear EDM's in ferroelectrics,[13] and for nuclear and electronic EDM's and T-odd e-N interactions in ^{3}He-A.[14]

Here we examine the simplest macroscopic P- and T-nonconservation experiment, magnetization of a paramagnet by applied electric field. Our proposal is related to those of Ignatovich and Vasilev and Kolycheva for ferromagnets; experiments on paramagnets appear to be simpler, however, and in particular are less prone to artifact, as discussed below. Our estimates of noise both in existing magnetometers and in the sample itself suggest that these experiments can push several orders of magnitude beyond current limits on the electron EDM. Before attempting to observe EDM-related effects in solids, however, we must convince ourselves of four points:

(1) That systems can be found in which intrinsic EDM's are not screened out at the atomic level; the basic problem was pointed out by Schiff.[15] Insofar as the atom can be described by nonrelativistic quantum mechanics, the nucleus can be treated as a point particle, and certain magnetic moment effects can be ignored, Schiff's theorem states that the electric dipole moment of an atom induced by intrinsic EDM's is zero to first order. Fortunately, relativistic effects are significant in heavy atoms, where the electronic EDM can actually be enhanced rather than screened.[10,16]

(2) That chemical bonding does not obliterate the atomic EDM. Clearly if the free spins which generate the atomic dipole moment are paired by bonding then the net EDM will be zero. On the other hand, if the heavy atom retains a net spin after bonding, as in the rare-earth and iron-group salts, then the crystalline environment exerts a relatively small effect on the total EDM.[10]

(3) That an external electric field will actually be felt by atoms in the interior of the sample. For dielectric materials, this is not a problem; because of the Lorentz correction the internal field can even exceed the external field.

(4) That the T-nonconserving effects which we seek to observe cannot be mimicked by effects associated with broken inversion symmetry in the crystal. Magnetization of the sample in response to an electric field and in the absence of an external magnetic field reflects an effective interaction energy $\sim \mathbf{E}\cdot\mathbf{B}$ which is

forbidden by T-invariance independent of crystal structure. An interaction energy linear in the magnetization could arise, however, if the material itself spontaneously breaks T invariance or as a "dirt effect" due to residual magnetic fields. The first effect can be eliminated if we use samples which exhibit no magnetic order. In the latter case, an effect quadratic in the total magnetic field would contain cross terms linear in the magnetization. Terms of the dangerous type, linear in the electric and quadratic in the magnetic field, necessarily also do not conserve parity and are related to the existence of electric-field–dependent g-value shifts in magnetic-resonance experiments.[17] These interactions are of the form $\sim \mu_e \mathbf{S} \cdot (\mathbf{T} \cdot \mathbf{E}) \cdot \mathbf{H}$, where $\mathbf{S}$ is the spin vector, μ_e is the electron magnetic moment and $\mathbf{T}$ is a third-rank tensor which expresses the dependence of the (second-rank) g-tensor on the electric field. To detect a T-nonconserving interaction $\sim d_e \mathbf{S} \cdot \mathbf{E}$ we thus require $\mu_e |\mathbf{T}||\mathbf{H}| << d_e$.

For inversion-symmetric crystal structures—including EuS discussed below—the tensor $\mathbf{T}$ vanishes identically. Point defects and substitutional impurities spoil the inversion symmetry of nearby sites, but most of these sites can be paired off as inversion images of one another so that the net linear electric field effect is zero.[17] Similar considerations apply to edge dislocations, but now one finds a single plane of sites which are inversion asymmetric and uncompensated by inversion images. Although these considerations are difficult to quantify without direct experimental evidence, note that even at very asymmetric sites rare-earth and transition-metal ions typically have[17] $|\mathbf{T}| \sim 10^{-7}$ cm/V so that in low-field ($H \lesssim 10^{-8}$ G) environments[18] these effects can mask $d_e \sim 10^{-27}$ $e \cdot$cm only if "very asymmetric sites" are at an effective concentration of $\gtrsim 1{:}10^4$; if these sites are $\sim 1{:}10^9$ then they are unimportant in any forseeable EDM search. With reasonable dislocation densities[19] and the compensation of inversion image sites it is plausible that these effective concentrations can be reached, but this should be checked by direct measurements of the electric-field–induced g-value shifts in the samples of interest.

These considerations suggest that candidate materials are insulating compounds of heavy elements with unfilled d or f shells at temperatures above the transitions to magnetic order in inversion-symmetric crystals. The experiment which we envision is then an analog of conventional paramagnetic susceptibility measurements in which a field is applied and the magnetization detected by some means. Limits to the detection of magnetization are imposed both by detector noise and by thermal fluctuations in magnetization which are intrinsic to the sample, while the magnitude of the signal is controlled by the effective atomic EDM and by the true magnetic susceptibility.

We begin by estimating the sensitivity of the experiment as limited by thermal noise in the sample itself. The quantity to be measured is the average magnetization M of the sample. If all of the magnetic moments in the sample are associated with electron spin rather than orbital angular momentum, then an electric field E on each atom is equivalent to a magnetic field $H_{\mathrm{eff}} = d_{\mathrm{eff}} E/\mu_e$, where d_{eff} is the effective atomic EDM per electron spin. Then $M = \chi E d_{\mathrm{eff}}/\mu_e$, where χ is the conventional magnetic susceptibility. The fluctuations in M, measured through a bandwidth Δf around ω, are given by[20]

$$V\langle(\delta M)^2\rangle = 4k_{\mathrm{B}}T\omega^{-1}[\mathrm{Im}\chi(\omega)]\Delta f \sim 4k_{\mathrm{B}}T\chi\tau\Delta f, \quad (1)$$

where V is the sample volume, τ is the paramagnetic relaxation time in the Debye approximation $[\chi(\omega) \sim \chi(1 - i\omega\tau)^{-1}]$, and the measurement is made at low frequencies ($\omega\tau << 1$). These fluctuations impose a thermal noise limit on the detection of d_{eff},

$$(d_{\mathrm{eff}})^2_{\mathrm{TN}} = 4k_{\mathrm{B}}T\frac{\tau\Delta f}{V\chi}\frac{\mu_e^2}{E^2} \sim (2.7\times10^{-29}\ e\cdot\mathrm{cm})^2 \frac{T}{10\ \mathrm{K}} \frac{\tau}{10^{-10}\ \mathrm{s}} \frac{\Delta f}{10^{-4}\ \mathrm{Hz}} \frac{10\ \mathrm{cm}^3}{V} \frac{1}{\chi} \left[\frac{10^5\ \mathrm{V/cm}}{E}\right]^2. \quad (2)$$

SQUID magnetometry provides the most sensitive technique currently available to detect very small magnetic fields. Clarke *et al.*[21] have discussed the optimal sensitivity of an untuned dc SQUID magnetometer. For a source coil of length 20 cm and radius 5 cm, they find that the field noise is

$$\langle(\delta B)^2\rangle \sim (2\times10^{-12}\ \mathrm{G})^2[\Delta f/(1\ \mathrm{Hz})].$$

Furthermore, δB scales with source coil volume (for fixed shape) as $\delta B \sim V^{-1/2}$. Comparing this to $B = 4\pi M = 4\pi E\chi d_{\mathrm{eff}}/\mu_e$, we find the instrumental detection limit

$$(d_{\mathrm{eff}})^2_{\mathrm{SQUID}} \sim (8.3\times10^{-28}\ e\cdot\mathrm{cm})^2[\Delta f/(10^{-4}\ \mathrm{Hz})][(10\ \mathrm{cm}^3)/V]\chi^{-2}[(10^5\ \mathrm{V/cm})/E]^2. \quad (3)$$

Note that the relative significance of thermal and instrumental noise is material and temperature dependent.

These results suggest that EDM searches should be carried out at a temperature just above the Curie point so as to achieve a large χ. There are, however, two considerations which may limit this diverging sensitivity. First, the relaxation time τ also diverges as the Curie temperature is approached, so that

$$(d_{\rm eff})^2_{\rm TN} \sim |T-T_c|^{\Delta-\gamma},$$

where Δ and γ are the critical exponents for τ and χ, respectively. Theoretical understanding of the dynamic exponent Δ is not sufficient to predict reliably whether sensitivity to the EDM is enhanced or degraded as $T \to T_c$, and so empirical results must be used on a case by case basis.

Second, the *effective* susceptibility $\chi_{\rm eff}$ of a sample is limited by its demagnetizing field, which is geometry dependent; as χ diverges, $\chi_{\rm eff}$ approaches a constant. In order to take advantage of large χ near the Curie point it is thus necessary to find an appropriate experimental geometry. One possibility is to place a low-aspect-ratio (length << diameter) sample (which is between nonsuperconducting capacitor plates) in a high-aspect-ratio superconducting solenoid consisting of discrete loops which float freely at different voltages. This geometry allows a large uniform electric field while simulating the magnetic properties of a very long sample so as to reduce the importance of demagnetization.

As a candidate material we consider EuS just above its Curie point at $T_c = 16.56$ K. Direct measurements[22] establish $\chi = 5$ at $T = T_c + 1$ K, where Debye relaxation is observed to hold and $\tau = 2\times10^{-10}$ s. The ratio $d_{\rm eff}/d_e$ is estimated to be 0.5 for the Eu^{++} ion,[6] and with a dielectric constant[23] $\epsilon = 11$ the effective electric field is $E = (\epsilon+2)E_{\rm ext}/3$, where $E_{\rm ext}$ is the externally applied field. We obtain the limits to detection of the electron EDM:

$$(d_e)^2_{\rm TN} \sim (7.4\times10^{-30}\ e\cdot{\rm cm})^2[(10\ {\rm cm}^3)/V][\Delta f/(10^{-4}\ {\rm Hz})][10^5\ {\rm V/cm})/E_{\rm ext}]^2, \quad (4a)$$

$$(d_e)^2_{\rm SQUID} \sim (7.7\times10^{-29}\ e\cdot{\rm cm})^2[(10\ {\rm cm}^3)/V][\Delta f/(10^{-4}\ {\rm Hz})][(10^5\ {\rm V/cm})/E_{\rm ext}]^2. \quad (4b)$$

For these conditions the SQUID limit is apparently more serious.

The current limit on the electron EDM is, as noted above, $d_e \le 10^{-24}\ e\cdot$cm. This certainly can be surpassed by our proposed experiment if the performance of Eqs. (4) can be reached. While sample volumes of several cubic centimeters are quite reasonable and electric fields of $\sim 10^5$ V/cm are routinely applied to dielectric materials, the use of a narrow bandwidth requires some discussion. If this bandwidth were centered at zero frequency our measurement would be swamped by $1/f$ noise; to avoid this problem we must make the measurement at $f > 10^{-2}$ Hz, the approximate corner frequency of a dc SQUID.[24] But if we apply an electric field E at frequency f an unwanted magnetic field $B \sim (fl/c)E$ is induced (l is a typical sample dimension); this field will magnetically polarize the sample and generate an artifactual signal. Even at $f \sim 10^{-2}$ Hz and $l \sim 1$ cm, $E \sim 10^5$ V/cm implies $B \sim 10^{-10}$ G which is enormous on the scale of interest. Fortunately this artifact is both spatially and temporally orthogonal to the true signal.

If the apparatus has perfect cylindrical symmetry, with the electric field applied and the magnetization detected along the cylinder's axis, the magnetic field is oriented radially and generates no observable signal. In practice the magnitude of the artifact will be reduced by the geometrical tolerances in the construction of the apparatus, with $1{:}10^3$ being reasonable for samples of centimeter dimensions. Furthermore, the electric and magnetic fields are in quadrature, so that with phase-sensitive (lock-in) detection the effective magnitude of the artifact decreases with integration time and is in fact frequency independent. In a bandwidth $\Delta f \sim 10^{-4}$ Hz surrounding $f \sim 10^{-2}$ Hz, this provides another factor of $\sim 10^2$. Finally, the small remaining artifact ($\sim 10^{-15}$ G, comparable to or smaller than the SQUID noise) can be exactly subtracted by use of frequency modulation: Since the magnetic field is proportional to the measurement frequency while the electric field is not, modulation of this frequency allows us to observe the artifact independent of the signal. We conclude that ac measurements can be done without generating unacceptable artifacts, and that in this way the $1/f$ noise barrier to narrow bandwidth detection can be overcome.

One further concern is that application of large ac electric fields may produce spurious signals at the detector and associated electronics. These (and other) artifacts can be avoided in a multiple modulation scheme, where in addition to the frequency-modulated ac electric field, the temperature of the sample is also slowly modulated. Since $T \sim T_c$, relatively small temperature modulations will change χ by a large amount. With lock-in detection of these nonlinear signal variations the experiment becomes sensitive only to signals that are proportional to both the electric field and the magnetic susceptibility. This modulation requires that the sample temperature is part of a feedback loop, and has the further advantage of bringing potentially serious temperature fluctuations down to a level limited by thermometer noise at the measurement frequency.

Multiple modulation, the inversion symmetry of the sample's crystal structure, and the availability of low-field environments make it plausible that the optimal performance of Eqs. (4) can be reached in a real experiment. It may be possible to go much further:

(1) We can simply try to scale the experiment described above. A bandwidth $\Delta f \sim 10^{-4}$ Hz corre-

sponds to an integration time of ~ 1 h, which is quite modest. Judging by the long-term cryogenic experiments run as magnetic monopole searches[25] or the Stanford ^{3}He EDM search,[26] integration times of several months should be feasible, reducing the detection threshold by a factor of 30 or more.

(2) As Clarke *et al.*[21] write, "Since one can, in principle, make the coil volume arbitrarily large, the ultimate sensitivity appears to be fundamentally limited only by Dewar size, assuming that spurious noise sources can be made negligible." If it were possible to do long-term experiments on several liters of material, sensitivity would extend below $d_e \sim 10^{-31}$ $e \cdot$cm.

(3) As we approach the Curie point in EuS, the relevant critical exponents[22] are $\Delta = 1.26 \pm 0.03$ and $\gamma = 1.41 \pm 0.01$; for the experimental sample of Ref. 22 the Debye approximation was valid only for $T > T_c + 0.3$ K. The effective thermal and SQUID noise levels scale as $\tau\chi^{-1}$ and χ^{-2}, respectively, so that as $T \rightarrow T_c$ the thermal noise limit changes very slowly while the SQUID limit decreases quite rapidly. If we operate at $T_c + 0.3$ K rather than $T_c + 1$ K as above, thermal and SQUID limits are essentially equal and we gain another order of magnitude in sensitivity to d_e. To achieve this improvement, however, one must still overcome the demagnetization effects discussed above.

Obviously each of these improvements to the basic experiment presents practical problems, as noted. Our intention here is only to indicate the possibilities for exploring well beyond $d_e \sim 10^{-27}$ $e \cdot$cm.

In summary, macroscopic T-nonconservation experiments on EuS using existing SQUID technology can clearly surpass present limits on the electron EDM. Small-scale experiments should open a 3-order-of-magnitude window beyond the current limit, at which point these measurements may compete with the neutron EDM for absolute sensitivity to T-nonconserving interactions. Large-scale, long-term searches for macroscopic T nonconservation have the potential for still greater sensitivity, and should be able to test nearly all available theoretical predictions.

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[1]J. H. Christenson, J. Cronin, V. L. Fitch, and R. Turlay, Phys. Rev. Lett. **13**, 138 (1964).

[2]K. R. Schubert, J. C. Chollet, J. M. Gaillard, M. R. Jane, T. J. Ratcliffe, and J. P. Repellin, Phys. Lett. **31B**, 662 (1970); R. C. Casella, Phys. Rev. Lett. **21**, 1128 (1968), and **22**, 554 (1969).

[3]For reviews see N. Ramsey, Annu. Rev. Nucl. Part. Sci. **32**, 211 (1982); R. Gollub and J. M. Pendlebury, Contemp. Phys. **13**, 519 (1972).

[4]Ramsey, Ref. 3.

[5]The most recent published result is T. G. Vold, F. J. Raab, B. Heckel, and E. N. Fortson, Phys. Rev. Lett. **52**, 2229 (1984).

[6]I. B. Khriplovich and A. R. Zhitinsky, Phys. Lett. **109B**, 490 (1982).

[7]A. Zee, Phys. Rev. Lett. **55**, 2379 (1985).

[8]M. B. Gravela and H. Georgi, Phys. Lett. **119B**, 141 (1982).

[9]W. M. Fairbank, unpublished, cited by L. I. Schiff, Phys. Rev. **132**, 2194 (1963).

[10]V. K. Ignatovich, Zh. Eksp. Teor. Fiz. **56**, 2019 (1969) [Sov. Phys. JETP **29**, 1084 (1969)].

[11]B. V. Vasilev and E. V. Kolycheva, Zh. Eksp. Teor. Fiz. **74**, 466 (1978) [Sov. Phys. JETP **47**, 243 (1978)].

[12]A. J. Leggett, Phys. Rev. Lett. **39**, 587 (1977).

[13]A. J. Leggett, Phys. Rev. Lett. **41**, 586 (1982).

[14]I. B. Khriplovich, Pis'ma Zh. Eksp. Teor. Fiz. **35**, 392 (1982) [JETP Lett. **35**, 485 (1982)].

[15]Schiff, Ref. 9.

[16]P. G. H. Sandars, J. Phys. B **1**, 499, 511 (1968).

[17]W. B. Mims, *The Linear Electric Field Effect in Paramagnetic Resonance* (Clarendon, Oxford, 1976).

[18]B. Cabrera, thesis, Stanford University 1975 (unpublished).

[19]C. Kittel, *Introduction to Solid State Physics* (Wiley, New York, 1976).

[20]L. D. Landau and E. M. Lifshitz, *Statistical Physics* (Pergamon, Oxford, 1977).

[21]J. Clarke, C. D. Tesche, and R. Giffard, J. Low Temp. Phys. **37**, 405 (1979). Subsequent improvements in SQUID noise levels have been reported—see J. Clarke, in *Advances in Superconductivity*, edited by B. Deaver and J. Ruvalds (Plenum, New York, 1985)—but it is not completely clear how these translate into magnetometer sensitivity in the experiment discussed here.

[22]M. Shiino and T. Hashimoto, J. Phys. Soc. Jpn. **45**, 22 (1978).

[23]P. Wachter, in *Handbook on the Physics and Chemistry of Rare Earths*, edited by K. A. Gschneider, Jr., and L. Eyring (North-Holland, Amsterdam, 1979).

[24]J. Clarke, in *Superconducting Quantum Interference Devices and Their Applications*, edited by H. D. Hahbolm and H. Lübbig (Walter de Gruyter, New York, 1977).

[25]See for example S. Bermon, P. Chaudhari, C. C. Chi, C. D. Tesche, and C. C. Tsuei, Phys. Rev. Lett. **55**, 1850 (1985), and references therein.

[26]W. M. Fairbank, private communication.