Unique Measurement of Time Reversal Symmetry Breaking with Optical Kerr Rotation

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I. INTRODUCTION

Recently, there has been controversy surrounding the apparent measurement of finite polarization rotation, also known as the magneto-optical polar Kerr Effect, in optical reflection measurements of materials which break microscopic time reversal symmetry. This argument applies if the linear response regime, and only fails for nonlinear effects and, in some cases, non-equilibrium effects. Recent measurements with a modified Sagnac Interferometer have found finite Kerr rotation in a variety of superconductors. Our result implies that the Sagnac Interferometer only measures time reversal symmetry breaking, and, thus, time reversal symmetry is broken in these materials.

As the Kerr angle is \( \theta_K = \frac{1}{2} (\text{arg} R_{++} - \text{arg} R_{--}) \), then it will be zero when the material preserves time reversal symmetry. While his argument is satisfactory, it is deserving of a more rigorous discussion.

We begin as Halperin does. Consider a general measurement of the reflection amplitudes where the pair of sources, each collocated with a detector, are positioned arbitrarily with respect to a sample and each other. Let the sources be of arbitrary shape, but emit light which, in the absence of all other sources or scatterers, appears as a circularly polarized plane wave at infinite distance. In the presence of scatterers, the emitted field may still be described as having a circular polarization state \( \pm \) near the source, if not as a plane wave. We consider the experiment where light of the + polarization state is emitted at a source located at \( \mathbf{r}_1 \) and the + component of the reflected wave is measured at a detector at \( \mathbf{r}_2 \). Let, also, light of the \( \pm \) polarization state, be sourced at \( \mathbf{r}_2 \) and the \( \mp \) component of the polarization be measured at \( \mathbf{r}_1 \). This is accomplished if the collocated detectors are such that they signal the arrival of a photon in the time-reverse of the quantum optical state initially formed at the source. Again, the Kerr angle is the measured difference in complex arguments of the two propagation amplitudes. In the limit of \( \mathbf{r}_1 \rightarrow \mathbf{r}_2 \rightarrow \infty \), the measured reflection amplitudes are the same as \( R_{++} \) and \( R_{--} \) described by Halperin.

We will demonstrate that when the instrumentation and the sample consist of materials which are all time reverse symmetric, the electromagnetic propagation amplitude from \( \mathbf{r}_1 \) to \( \mathbf{r}_2 \) will always be identical to that for propagation from \( \mathbf{r}_2 \) to \( \mathbf{r}_1 \). It then follows that the Kerr angle will also be zero when there is time reversal symmetry, and that broken mirror symmetry, alone, can not give rise to Kerr rotation.

II. PROPAGATORS FOR OPTICAL MEASUREMENTS

Photon Green’s functions describe optical measurements. In the macroscopic limit, the light emitted from a source and measured by a detector is modeled by the
The retarded Green’s function for the Macroscopic Maxwell’s equations:
\[
\begin{align*}
\nabla \times E &= -\frac{1}{c} \frac{\partial}{\partial t} B \\
\nabla \times H &= \frac{1}{c} \frac{\partial}{\partial t} D + i \omega J \\
\nabla \cdot B &= 0 \\
\n\nabla \cdot D &= \rho_f
\end{align*}
\]

Where \( B = \nabla \times A \) and \( E = -\frac{1}{c} \frac{\partial}{\partial t} A \) in radiation gauge. At optical frequencies, it is sufficient to describe the material’s response with just a dielectric susceptibility function, \( \chi(t_2, r_2, t_1, r_1) \). The retarded Green’s function, \( G^{\text{ret}} \), relates the source current, \( J \) to the macroscopic vector potential, \( A \):
\[
A(t_2, r_2) = \int G^{\text{ret}}(t_2, r_2, t_1, r_1) J(t_1, r_1) dt_1 \, dr_1
\]

Precise statements of the symmetries of the electromagnetic field and its measurement entail that the reflection amplitudes are considered quantum mechanically. The quantum electrodynamic field measured at \( (t_2, r_2) \) by a point dipole detector, aligned to the \( \mu \) linear polarization state, will be \( A_\mu(t_2, r_2) |0\rangle \), where \( A_\mu(0, r) = A^\dagger_\mu(0, r) \) is the position-space field operator and \( |0\rangle \) is the vacuum state. Likewise, supposing a point-like dipole source creates a \( \nu \) linearly polarized photon at \( (t_1, r_1) \), then the quantum field it initially forms will be \( A_\nu(t_1, r_1) |0\rangle \). For \( t_2 > t_1 \), the amplitude for free-space propagation between the source and receiver is given by:
\[
\langle 0 | A_\mu(t_2, r_2) \hat{A}_\nu(t_1, r_1) |0\rangle = \delta_{\mu\nu} \frac{\delta(t_2 - t_1 - \frac{1}{c} |r_2 - r_1|)}{4\pi |r_1 - r_2|}
\]

Squared, this is the transition probability for the detection of a photon at time \( t_2 \) given its creation at \( t_1 \).

When the sources are of a single frequency \( \omega \), the phase delay, as used to define the Kerr angle, is the complex argument of the propagator in the frequency-position domain:
\[
G^{\text{ret}}(\omega; r_2, r_1) = \int G^{\text{ret}}(t, r_2, t_0, r_1) e^{i\omega t_0} dt
\]

When the light is interacting with matter, then to lowest order, the linear response of the macroscopic field at the detector, \( A(t, r_2) \), for \( r_2 \) outside of the material, to an optical source at \( r_1 \), also outside of the material, is given by Equation. The retarded Green’s function is obtained by complex conjugating the negative frequency part of the following time-ordered propagator:
\[
G^{\text{ret}}_{\mu\nu}(t_2, r_2, t_1, r_1) = \langle g | T[\hat{A}_\mu(t_2, r_2) \hat{A}_\nu(t_1, r_1)] | g \rangle
\]

where \( T \) is the time-ordering operator for photons: \( T[\hat{A}_\mu(t_2, r_2) \hat{A}_\nu(t_1, r_1)] = \theta(t_2 - t_1) \hat{A}_\mu(t_2, r_2) \hat{A}_\nu(t_1, r_1) + \theta(t_1 - t_2) \hat{A}_\nu(t_1, r_1) \hat{A}_\mu(t_2, r_2) \). We choose to focus on the time-ordered propagator to emphasize that it is readily calculable from quantum perturbative methods.

The expectation value is taken with respect to the many-body ground state, \( |g\rangle = \lim_{t \to -\infty} \hat{g}^\dagger(t) |0\rangle \) of the whole system. This ground state includes the material, the environment and any instrumentation. If the system is at finite temperature, then a Boltzmann weighted sum of propagators, evaluated with respect to the stationary states of the system is used in lieu of the above. In this way, even incoherent optical sources may be described.

In assuming that the measurement is described exactly by Equation it is implied that the source is the perturbation to the full Hamiltonian of the world, \( H \), which describes the light, the material and the detectors. The perturbing source, \( J(t, r) \), is slowly turned on from zero at \( t = -\infty \) and slowly turned off at \( t = \infty \). It is also assumed that the sample, the source and receiver do not interact in any way other than by the scattered light; this is tantamount to requiring that the operators \( A_\mu(t, r_2) \) and \( A_\nu(t, r_1) \) commute with each other, and with \( \hat{g}(t) \) and \( \hat{g}^\dagger(t) \) at equal times. These are the same conditions requisite for application of the Kubo Formula, and results similar to those in the next section appear in many texts in connection with it.

III. THE RECIPROCITY THEOREM

We will prove that, if time reversal symmetry is respected, then no Kerr rotation is observed, by showing that this symmetry condition implies that the propagator for + polarized light traversing from \( r_1 \to r_2 \) and the propagator for − polarized light traversing from \( r_2 \to r_1 \) are identical. Of central importance is that the measurement is performed with collocated sources and detectors, which create or destroy photons in states which are the time-reverse of each other. This condition is clearly true for the two point-like dipole sources/detectors, located at \( r_1 \) and \( r_2 \), considered in this discussion. We later describe an example of how this is achieved in practice.

The anti-linear time reversal operator, \( T \), commutes with the Hamiltonian, \( H \): \( THT^{-1} = H \), but still inverts the time-evolution operator, \( T e^{-iHT}T^{-1} = e^{iHT} \), as well as anti-commutes with all other operator generators of motion. Its action on quantum states, \( u, v \), is, \( T|u\rangle = |v^*\rangle \), where the overbar represents the time-reversed state and * refers to the fact that the map is to the “complex conjugate Hilbert space,” where \( \langle u^*|v^*\rangle = \langle v|u \rangle \). The vector potential has odd time reversal parity, so \( T \hat{A}_\mu(0, r)T^{-1} = -\hat{A}_\mu(0, r) \). Since \( \hat{A}_\mu(t, r) = e^{iHT} \hat{A}_\mu(0, r) e^{-iHT}, \) then \( T \hat{A}_\mu(t, r)T^{-1} = -\hat{A}_\mu(-t, r) \).

It follows that:
\[
\langle g | T[\hat{A}_\mu(t_2, r_2) \hat{A}_\nu(t_1, r_1)] | g \rangle
\]

\[
\langle g | T[\hat{A}_\mu(t_2, r_2) \hat{A}_\nu(t_1, r_1)]T\hat{A}_\mu(t_1, r_1)T^{-1} | g \rangle
\]

\[
\langle g | T[\hat{A}_\nu(t_2, r_2) \hat{A}_\mu(t_1, r_1)] | g \rangle
\]

\[
\langle g | T[\hat{A}_\nu(t_2, r_1) \hat{A}_\mu(t_1, r_2)] | g \rangle
\]
Where the last equality follows from time-translation symmetry. It is then the case that if the ground state of the material is time reversal symmetric, \( |g\rangle = |g\rangle \), that

\[
G^R_{\mu\nu}(t_2,t_2,t_1,r_1) = G^R_{\nu\mu}(t_2,t_1,r_1,t_2)
\]

(4)

There is a similar derivation of this symmetry for the retarded propagator, or else, it is obtained from analytic continuation of the above.

We refer to this result as the “Reciprocity Theorem” or “reciprocity” symmetry, and it is only satisfied when the ground state possesses microscopic time reversal symmetry. Again, the restriction to point-like dipole sources is unnecessary, as an extended source is described by integrating \( r_1 \) and \( r_2 \) over the respective volumes. Linear absorption in the sample is inconsequential; while the transition, as we evaluate expectation values of the propagator manifestly breaks time reversal symmetry, \( \bar{A} \), which break time-translation symmetry and invalidate the last step in \( \mathbf{3} \). In other words, Equation 5 fails when microscopic time reversal symmetry is broken, but does not distinguish between systems in which it is broken due to a phase of matter which arises from spontaneous symmetry breaking, or from an external forcing, as in the Spin Hall Effect, where an applied current results in an unbalanced population of spins. Likewise, there might be a highly excited state of a material which breaks mirror symmetry and emits radiation, as it relaxes, symmetrically in the two circular polarization states, again, leading to an unbalanced spin population. If these nonequilibrium systems are steady state, then there will be a density matrix, \( \bar{\rho} \), which is not Boltzmann and is used to evaluate Equation 2. Unless this density matrix manifestly breaks time reversal symmetry, \( [\bar{T},\bar{\rho}] \neq 0 \), then the measurement will satisfy reciprocity and there can be no Kerr rotation.

IV. CONCLUSION

In proving Equation 5 we have dispelled some incorrect ideas recently promulgated as well as affirm and
clarify the work of a number of studies\cite{20,20,74–77}. To summarize: (1) Kerr rotation may only arise from microscopic time reversal symmetry breaking. This symmetry breaking may occur either through spontaneous symmetry breaking, or by nonequilibrium processes. Circular dichroism, as measured by the difference in reflected powers of two beams of normally incident and oppositely circular polarized light, may also only arise from microscopic time reversal symmetry breaking. Corollary to this, optically active materials, such as those with a k-linear susceptibility or any other form of mirror symmetry breaking, can not give rise to the magneto-optical Kerr effect, as they are time reversal symmetric. (2) The proof above coincides with Onsager’s relations and the Electromagnetic Reciprocity Theorem, and all three will fail only when microscopic time reversal symmetry is broken. The theorems do not apply for nonlinear response, however one may always tell if the sample or instrumentation is responding nonlinearly with evidence of intensity dependent observables or an alteration in the reflected frequency spectrum. There are nonlinear effects which are intensity-independent and only alter the spectral content, such as spontaneous Raman shifts or spontaneous parametric photon down-conversion, although these effects are incoherent and yield a random phase delay.

A common source of confusion impedng the acceptance of these arguments have been calculations of Kerr rotation when using the constitutive relations for optical activity given by \( \mathbf{B} = \mathbf{H} \) and \( \mathbf{D} = \varepsilon^0(\mathbf{r})\mathbf{E} + \gamma(\mathbf{r})\nabla \times \mathbf{E} \) or \( \mathbf{D} = \varepsilon^0(\mathbf{r})\mathbf{E} + \nabla \times (\gamma(\mathbf{r}))\mathbf{E} \), where \( \varepsilon^0 \) is the isotropic permittivity and \( \gamma \) is the spatially dependent gyrotropic parameter.\cite{20,20,74–77} These relations are fine when the material is homogeneous, however, when there is a surface or spatial inhomogeneity, these constitutive relations do not satisfy Onsager’s relations,\cite{24,24,25} which means they do not explicitly satisfy time reversal symmetry. Furthermore, in lossless media, they don’t respect Poynting’s Theorem\cite{20,20,24,25} or follow from a least action principle.\cite{22} For a material which is time-reverse symmetric, but breaks mirror symmetry one expansion for the permittivity tensor is:

\[
epsilon_{\mu\nu}(\omega, \mathbf{r}, \mathbf{r}') = \varepsilon^0_{\mu\nu}(\omega, \mathbf{r})\delta(\mathbf{r} - \mathbf{r}') - \gamma_{\mu\nu\lambda}(\omega, \mathbf{r}, \mathbf{r}')\partial_\lambda\delta(\mathbf{r} - \mathbf{r}')
\]

It is clear that \( \varepsilon_{\mu\nu}(\omega, \mathbf{r}, \mathbf{r}') = \varepsilon_{\mu\nu}(\omega, \mathbf{r}', \mathbf{r}) \) when \( \varepsilon^0_{\mu\nu}(\omega, \mathbf{r}) = \varepsilon^0_{\mu\nu}(\omega, \mathbf{r}) \) and \( \gamma_{\mu\nu\lambda}(\omega, \mathbf{r}, \mathbf{r}') = -\gamma_{\mu\nu\lambda}(\omega, \mathbf{r}', \mathbf{r}) \). Furthermore, for homogeneous media, \( \gamma_{\mu\nu\lambda} \) and \( \varepsilon^0 \) are constant; this gives a k-linear, bulk response. For isotropic media, one way \( \gamma_{\mu\nu\lambda} \) may be expressed in terms of \( \gamma \) is \( \gamma_{\mu\nu\lambda}(\omega, \mathbf{r}, \mathbf{r}') \equiv \varepsilon^0_{\mu\nu\lambda}(\gamma(\mathbf{r})\mathbf{r} + \frac{i}{2}\mathbf{r}) \). Regardless, because isotropic \( \gamma_{\mu\nu\lambda} \) must be symmetric in \( \mathbf{r} \) and \( \mathbf{r}' \), the same functional form for the correct constitutive relation is always obtained:

\[
\mathbf{D} = \varepsilon^0(\mathbf{r})\mathbf{E} + \frac{i}{2}\gamma(\mathbf{r})\nabla \times \mathbf{E} + \frac{i}{2}\nabla \times (\gamma(\mathbf{r})\mathbf{E}).
\]

This does not predict Kerr rotation.\cite{20,20,24,25,20,20,24,25}

The Sagnac Interferometer,\cite{20,20,74–77} the instrument used to measure the Kerr angle in Karapetyan et.al., being a unique test for reciprocity, only measures microscopic time reversal symmetry breaking. This is so because the interferometer measures the Kerr angle by interfering two beams of light made to reflect from the sample in a fashion such that the sourcing aperture for one beam is the receiving aperture for the other, and vice versa. The Sagnac Interferometer conveys light of two linear polarization states to the sample by a polarization maintaining, single mode optical fiber. The end-face of the fiber is an “aperture” for the two linear polarization states and the two modes which couple from free-space to the two fiber axes are the time-reverse of those two which are emitted from it. A quarter-wave plate with slow axis oriented at 45° with respect to the two polarization states emerging from the fiber axes is placed between the fiber end-face and the sample. The two orthogonal linearly polarized beams of light emitted from the fiber are transformed into opposite circularly polarized states after traversing the quarter-wave plate. The circularly polarized beams of light reflect from the sample into the same circular polarization states and will pass through the quarter-wave plate a second time, transforming back into orthogonal linear polarization states, but now rotated 90° from before. In this way, the beams couple from one axis of the fiber to the other and interfere at a polarizer, oriented at 45° with respect to both axes of the fiber, placed at the other end of the fiber optic cable. A lock-in amplifier technique recovers the Kerr angle from the interference intensity.\cite{22}

There exist other spurious optical paths, besides the two described above, which light may travel through the instrument and return to the detector; these do not affect the measurement. As an example, linear birefringence at the sample can partially reflect circularly polarized light into the reversed circular polarization state. This will cause light starting on one axis of the fiber, to reflect from the sample and back to the same axis of the fiber, after a double pass through the quarter-wave plate. However, because the fiber is highly birefringent and the diode light source has 8μm coherence length, only light which couples, after reflecting from the sample, between different axes in the fiber will traverse optical path lengths which differ by less than a coherence length and interfere coherently at the polarizer.\cite{22} Light which starts on one axis of the fiber and partially recouples, after a reflection off a birefringent sample, back to the same axis of the fiber will be incoherent with light that recouples to a different axis of the fiber, and not interfere.

The Reciprocity Theorem that we proved applies to the Sagnac interferometer exactly. The spatial filtering of the fiber ensures that the electromagnetic spatial modes which are sourced and received by the fiber are exactly the time-reverse of each other. Comparing the phase delays of light exchanged between the two fiber axes uniquely tests for microscopic time reversal symmetry breaking, not only in a sample being probed, but also within the optical components which make up the instrument itself. Misalignments or imperfect optical components will not introduce spurious signals, as they will have time reversal symmetric responses.
Because of the Reciprocity Theorem, the suggestion that the recent measurements of a Kerr effect stem from an equilibrium phase of matter, with mirror symmetry breaking and without time reversal symmetry breaking, can not be correct. Instead, the Reciprocity Theorem implies that either the ground state must break time reversal symmetry or the sample is in a highly nonequilibrium state which does as well. More tests are needed to determine if nonlinear effects are relevant.

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