Monitoring variations of refractive index via Hilbert–Schmidt speed and applying this phenomenon to improve quantum metrology

Seyed Mohammad Hosseiny¹, Hossein Rangani Jahromi²,* and Mahdi Amniat-Talab¹

¹ Physics Department, Faculty of Sciences, Urmia University, PO Box 165 Urmia, Iran
² Physics Department, Faculty of Sciences, Jahrom University, PO Box 74135111 Jahrom, Iran

E-mail: h.ranganijahromi@jahromu.ac.ir

Received 27 December 2022, revised 24 July 2023
Accepted for publication 14 August 2023
Published 29 August 2023

Abstract
In this paper, we investigate the role of the nonlinear response of a material to improve quantum metrology. We show that the collective optical behavior of an atomic ensemble can be applied to enhance frequency estimation through one of the atoms. In fact, the collective optical behavior of the atomic ensemble by analyzing the quantum information extracted from one of its elements. Moreover, we introduce Hilbert–Schmidt speed (HSS), an easily computable theoretical tool, to monitor the variations of linear as well as nonlinear refractive indices and evaluate the strength of the nonlinear response of optical materials. Furthermore, we illustrate that quantum Fisher information and HSS can efficiently detect negative permittivity and refractive index, which is of great importance from a practical point of view.

Keywords: non-linear optics, refractive index, Hilbert–Schmidt speed, quantum Fisher information

((Some figures may appear in colour only in the online journal)

1. Introduction
The field of quantum metrology [1–18] has provided valuable tools, such as the classical Fisher information and the quantum Fisher information (QFI) [19–27], for estimating the sensitivity of measurement devices and unknown parameters. Such analyses are usually focused on finding optimal measurement strategies, with respect to environmental parameters as well as initial conditions, to achieve measurement sensitivities better than the standard quantum limit [28]. However, the relationship between the optimal strategies for a measurement performed on a single atom through and the collective behavior of the atomic ensemble is rarely investigated.

One example of such phenomena, relevant for this context, is negative refraction.

When light propagates through a medium with negative values of both electric permittivity ($\epsilon < 0$) and magnetic permeability ($\mu < 0$), we encounter negative refraction. Such a medium, also known as a left-handed material or a metamaterial, was first proposed by Veselago in 1968 [29]. He showed that light in such a medium would have a negative refractive index, meaning that it would bend in the opposite direction to normal materials at an interface, and that it would also exhibit other unusual effects, such as reversed Doppler shift and Cherenkov radiation. These effects open up new possibilities for manipulating light, such as subwavelength imaging, cloaking, and superlensing [30, 31]. However, natural materials with negative values of both $\epsilon$ and $\mu$ are rare and only exist at certain frequency ranges. Therefore, researchers have...
explored alternative ways to achieve negative refraction, such as using materials with only one negative parameter (either \( \epsilon \) or \( \delta \)) [32–37], or using composite structures that combine different materials with positive and negative parameters. In this paper, we investigate how QFI and Hilbert-Schmidt speed (HSS), two important quantum metrological tools [10], can be used to detect and characterize these phenomena in atomic media with nonlinear optical responses.

Nonlinear optics [38] has been a rapidly developing field of science in recent years. It is based on the phenomena connected to the interaction of intense laser fields with matter. In fact, nonlinear optics focuses on the interactions of light with matter under conditions in which the nonlinear response of the atoms plays a significant role. These phenomena cover a broad range of applications [39], including spectroscopy [40, 41], telecommunications [42], optical data storage as well as processing [43–45], and quantum information technologies [46–48], especially quantum metrology [49].

One of the important parameters of interest to estimate in linear as well as nonlinear optics is the unknown frequency of a laser beam or radiation emitted in a spectroscopy experiment. Evaluating and controlling frequency or wavelength are vital in different situations to protect body organs or avoid damage to the materials. For example, light at a wavelength sufficiently long, not damaging biological materials, can be applied to gain a resolution requiring normally a much shorter wavelength [38]. Besides, the importance of the frequency dependence of the breakdown fields in different materials has been investigated [50–54]. Furthermore, the choice of frequency has a profound effect on the design of linear colliders [55]. Another phenomenon related to frequency is the dispersion, which is the frequency dependence of the refractive index of various materials in linear and nonlinear optics. A phase match based frequency estimation can be used to enhance the ranging precision of linear frequency modulated continuous wave radars [56]. More interestingly, the capability of decreasing the quantum noise in gravitational wave interferometers considerably depends on the frequency which should be detected [57]. Not only that, but quantum noise in the audio-band spectrum can be reduced by frequency-dependent squeezing schemes [58, 59]. These arguments motivate us to investigate frequency estimation and particularly explore how optical tools can be applied to achieve optimal measurement.

Our work is inspired by the recent paper [60], which introduces HSS as a powerful figure of merit for enhancing estimation of energy spectrum of a three-level atom. However, our work differs from the aforementioned reference in several aspects. First, we investigate the relationship between the collective behavior of an atomic ensemble and the specific behavior of its components, while [60] examines the behavior of a single three-level atom. Second, our paper focuses on the nonlinear response of the material to laser fields, while [60] focuses on the linear response. Third, our paper uses HSS to monitor variations in refractive index, while the aforementioned reference uses it to enhance estimation of energy spectrum. Therefore, our work extends the applicability of HSS to a broader range of problems in quantum metrology.

In this paper, we present a novel method for frequency estimation of input laser fields based on the optical behavior of an atomic ensemble. Unlike previous works that have used quantum metrology techniques [4, 61–72], our approach exploits the sensitivity of the HSS [73, 74] to the variations of the linear and nonlinear refractive indices of the optical material. We show that the HSS, which is a quantum statistical speed that can be easily computed, can be used to characterize metamaterials with negative permittivity and refractive index [75], which are important active fields for research in modern optics. We also demonstrate that our method can improve the frequency estimation by taking advantage of the phase matching condition of nonlinear optics [76], which depends on the refractive index of the sample being imaged. This condition ensures that the output wave maintains a fixed phase relation with respect to the nonlinear polarization and can extract energy most efficiently from the input waves. By monitoring the refractive index, we can optimize this condition and improve the accuracy of our frequency estimation.

The paper is structured as follows: in section 2, the concepts of QFI and HSS are explained. Then, after introducing two models in sections 3 and 4, we discuss how these concepts can be related to optical responses of the material. Finally, section 5 is devoted to summarizing and discussing the most important results.

It should be noted that, for plotting the figures, all parameters are nondimensionalized [77]. For quantities with the dimension of time and frequency, \( 1/\tilde{\omega} \) and \( \tilde{\omega} \) are set equal to 1. Here, \( \tilde{\omega} \) is an arbitrary value with the dimension of inverse time that is chosen to be \( 10^{15} \) Hz in our models (see table 1). Changing \( \tilde{\omega} \) does not affect the qualitative results.

### Table 1. Method of nondimensionalizing the parameters.

| Physical quantity | Nondimensionalized value | Real value (SI unit) |
|-------------------|--------------------------|----------------------|
| Time              | 1                        | \( \tilde{\omega}^{-1} = 10^{-15} \) s |
| Frequency         | 1                        | \( \tilde{\omega} = 10^{15} \) Hz |

### 2. Preliminaries

#### 2.1. QFI

The fundamental question when investigating the sensitivity of a quantum state with respect to other parameters is the following: by performing measurements on similar systems affected by some unknown parameter \( \eta \) (which may be, for example, a parameter quantifying the magnitude of a gravitational field or rotation, acceleration), how accurately can \( \eta \) be estimated? The answer is expressed by the quantum Cramer–Rao bound [78], dictating that the smallest resolvable change in the parameter \( \eta \) is given by...
where \( F_\eta \) denotes the QFI, which for pure states can be simply written as

\[
F_\eta = 4 \left| \langle \hat{\psi} | \hat{\psi} \rangle - |\langle \psi | \hat{\psi} \rangle|^2 \right|,
\]

which is an important quantifier of quantum statistical speed with respect to parameter \( \eta \). It is interesting to note that no diagonalization of \( d\rho(\varphi)/d\eta \) is required for HSS computation.

The HSS is known as an efficient tool for detecting non-Markovianity as well as improving quantum phase estimation in n-qubit open quantum systems. Generally speaking, because both HSS and QFI are quantum statistical speeds corresponding, respectively, to the Hilbert–Schmidt and Bures distances [78, 80], it is fascinating to investigate in detail how they can be related to each other in different scenarios.

Because the explicit expressions for the HSS and QFI in our models have cumbersome forms, they are not reported here.

3. Model I (four-level system)

We focus on a setup in which strong and weak laser fields \( E_s \) and \( E \) at frequencies, respectively, \( \omega_s \) and \( \omega \) are applied to a four-level atomic system in order to generate radiation at the sum frequency \( \omega = \omega_s + \omega \) (see figure 1) [38]. It is known that by allowing the field at frequency \( \omega_s \) to be a strong saturating field one, we can efficiently eliminate linear absorption at the \( a \rightarrow d \) transition frequency while gaining a large four-wave-mixing susceptibility. This kind of absorption elimination is called electromagnetically induced transparency (EIT). We assume that the conditions necessary to achieve the EIT are satisfied. It is emphasized that the nonlinear response can remain considerably large even when linear absorption at the output frequency vanishes in the EIT technique.

Our first goal is to investigate how the sum-frequency generation process is related to estimating frequencies of the driving laser fields and the quantum statistical speed. In particular, we study the nonlinear response leading to sum-frequency generation by analyzing its behavior via QFI and HSS computed for the frequencies of the driving fields.

The Hamiltonian of the atomic system can be split into two terms as \( H = H_0 + V(t) \) in which \( H_0 \) denotes the Hamiltonian of the free atom. Moreover, using the rotating-wave and electric-dipole approximations, one can see that \( V(t) \), designating the interaction energy of the atom with the externally driving radiation fields, can be expressed as

\[
V(t) = -\mu \left( E e^{i\omega t} + E^* e^{i\omega t} \right),
\]

where \( \mu \) represents the atomic dipole moment. Moreover, the Rabi frequencies \( \Omega_{ba}, \Omega_{cb}, \) and \( \Omega_{dc} \) are defined by

\[
\begin{align*}
\Omega_{ba} &= -\mu_{ba} E e^{-i\omega t}, \\
\Omega_{cb} &= -\mu_{cb} E e^{-i\omega t}, \\
\Omega_{dc} &= -\mu_{dc} E e^{-i\omega t},
\end{align*}
\]

where the notations \( \langle i | V | j \rangle \equiv V_{ij} \) and \( \langle i | \mu | j \rangle \equiv \mu_{ij} \) are adopted. In addition, the detuning factors

\[
\delta_1 = \omega - \omega_{ba}, \quad \delta_2 = \omega - \omega_{cb} \quad \text{and} \quad \Delta = \omega_s - \omega_{dc},
\]

in which \( \omega_{ba} = \omega_b - \omega_a, \omega_{cb} = \omega_c - \omega_b, \) and \( \omega_{dc} = \omega_d - \omega_c \), denote the transition frequencies, it should be noted that the energies are measured relative to that of the ground state \( |a \rangle \).

We should compute the wavefunction to find the response leading to sum-frequency generation. In the interaction picture, it can be expressed as [38]

\[
\psi(t) = C_a(t) |a \rangle + C_b(t) |b \rangle e^{-i\omega t} + C_c(t) |c \rangle e^{-i(\omega_2 + \omega) t} + C_d(t) |d \rangle e^{-i(\omega_s + \omega_2) t},
\]

obeying Schrödinger’s equation in the form

\[
i\hbar \frac{\partial |\psi\rangle}{\partial t} = H |\psi\rangle \quad \text{with} \quad H(t) = H_0 + V(t).
\]

Solving the Schrödinger equation perturbatively in terms of \( \Omega_{ba} \) and \( \Omega_{cb} \) but to all orders in \( \Omega_{dc} \) and taking \( C_i = 0 \) for the steady-state solution, we find that [38]

\[
\begin{align*}
C_a &= 1, \\
C_b &= -\Omega_{ba}/\delta_1, \\
C_c &= \Omega_{cb}/\delta_2, \\
C_d &= \left( \Omega_{dc}/\Delta \right) \delta_1^{\gamma}.
\end{align*}
\]
Now, calculating the induced dipole moment at the sum frequency, i.e., \( \mathbf{p} = \langle \psi | \mu | \psi \rangle \) one finds that the third-order nonlinear optical susceptibility is given by
\[
\chi^{(3)} = \frac{-N_{\text{had}} \mu_{dc} \mu_{ha}}{3\epsilon_0 \hbar \left( \delta_2 + \Delta + i \gamma_c \right) - |\Omega_{dc}|^2},
\]
where \( \epsilon_0 \) represents the vacuum permittivity and \( N \) denotes the number density of atoms. The effects of damping to this result can be added by replacing \( \delta_2 \) and \( \delta_2 + \Delta \) with, respectively, \( \delta_2 + i \gamma_c \) and \( \delta_2 + \Delta + i \gamma_d \), leading to [38]
\[
\chi^{(3)} = \frac{-N_{\text{had}} \mu_{dc} \mu_{ha}}{3\epsilon_0 \hbar \delta_1 \left( \delta_2 + i \gamma_c \right) \left( \delta_2 + \Delta + i \gamma_d \right) - |\Omega_{dc}|^2},
\]
where \( \gamma_d \) and \( \gamma_c \) represent the decay rates of the probability amplitudes to be in levels \( d \) and \( c \), respectively.

\( |\chi^{(3)}| \) quantifies the strength of the third-order nonlinear optical response of the material driven by the laser field. In the presence of strong laser field \( E_s \), the refractive index, experienced by a weak wave, can be written as [81] \( n = n_0 + n_2 I \), in which \( n_0 \) denotes the usual (i.e. low-intensity or linear) refractive index. Moreover, \( n_2 \), called the nonlinear refractive index, characterizes the strength of the optical nonlinearity induced by the strong field. If the system exhibits negligible absorption, it is given by \( n_2 = \frac{3}{\omega_0 c} |\chi^{(3)}| \), in which \( c \) denotes the speed of light. In [82] the general connection between \( n_2 \) and \( |\chi^{(3)}| \) has been discussed. In addition, \( I = 2n_0 \epsilon_0 |E_s|^2 \) represents the time-averaged intensity of the strong wave. Furthermore, the linear refractive index \( n_0 \) is connected with the linear susceptibility \( \chi^{(1)} \) and linear dielectric constant \( \epsilon^{(1)} \) via
\[
n_0 = \sqrt{\epsilon^{(1)}} = \sqrt{1 + \chi^{(1)}}.
\]

The imaginary part of the refractive index characterizes the absorption of radiation which occurs due to the transfer of population from the atomic ground state to some excited state. Strictly speaking, the low-intensity normal (i.e. linear) absorption coefficient can be represented in terms of the linear susceptibility \( \chi^{(1)} \) through
\[
\alpha_0 = \chi^{(1)*} \omega/c,
\]
where the real and imaginary parts of the linear susceptibility have been defined as \( \chi^{(1)} = \chi^{(1)} + i \chi^{(1)*} \). The absorption coefficient of many material systems decreases when measured using high laser intensity. Considering an incident laser radiation with intensity \( I \), one finds that the (high-intensity) absorption coefficient is given by \( \alpha(I) = \alpha_0 / (1 + I/I_s) \) in which \( I_s \) denotes the saturation intensity for which \( \alpha(I_s) = \alpha_0 / 2 \). However, in this paper, we investigate a weak laser field that is not strong enough to considerably correct its own absorption. Nevertheless, the strong laser field \( E_s \), coupling the two upper energy levels, can affect the absorption of the weak field \( E \) through another mechanism called EIT.

### 3.1. Relation between strength of non-linear response and quantum statistical speeds

In this section, we reveal important relationships between the non-linear response of the material to the optical driving fields and the quantum statistical speeds of a single four-level atom located inside the material. Computing the QFI and HSS versus frequency of the weak or strong field, we see that they exhibit completely similar qualitative behaviors. We also show that they can be used as efficient tools to measure the degree of the non-linear response of the material to the driving laser fields.

#### 3.1.1. Quantum statistical speeds with respect to \( \omega_s \)

Figure 2(a) compares the variations of the QFI, HSS, computed with respect to \( \omega_s \), and \( |\chi^{(3)}| \) versus \( \omega_{dc} \). It is clear that if \( \delta_2 = 2\omega - \omega_{0a} = 0 \), the best estimation occurs when \( \Delta_1 = \omega_s - \omega_{dc} = 0 \), i.e. \( \omega_{dc} = \omega_s \). Therefore, the most precise frequency estimation of the strong field is obtained when it is resonant with the transition frequency \( \omega_{ha} \).

Now, we investigate how control parameters \( \omega \) and \( \Omega_{dc} \) should be fixed to achieve the optimal estimation of \( \omega_s \). Figure 2(b) demonstrates that the maximum point of the QFI rises when \( \delta_1 = \omega - \omega_{0a} = 0 \), i.e. \( \omega = \omega_{ha} \). Hence, the weak laser field should be tuned to resonance with the transition frequency \( \omega_{ha} \) to achieve the most accurate estimation.

In figures 2(a)–(c) we observe that the variations of the QFI and HSS, calculated with respect to \( \omega_s \), are in perfect agreement with each other. In addition, HSS, \( \langle F_{\omega} \rangle \), can be used to predict the strength of the nonlinear response. In fact, the point at which the HSS is maximized versus each of the parameters: \( \omega_{dc}, \omega, \Omega_{dc} \) exactly reveals the value for which the strongest nonlinear response of the medium occurs. Moreover, because the maxima of the QFI and \( |\chi^{(3)}| \) coincide, the nonlinear response can be considered as a source for achieving the optimal frequency estimation of the strong field.

It is also found that an increase in the Rabi frequency \( \Omega_{dc} \) first enhances the frequency estimation until the maximized nonlinear response of the medium is achieved (see figure 2(c)). Then, we see that an increase in the \( \Omega_{dc} \) results in suppression of the nonlinear response as well as the accuracy of the estimation.

#### 3.1.2. Quantum statistical speeds with respect to \( \omega_s \)

Figure 3 demonstrates that computation of the QFI and HSS with respect to \( \omega_s \) can provide us with valuable information on the nonlinear response of the material. Investigating their variations versus \( \omega_{dc} \) or \( \omega_s \), we see their common minimum point exactly coincide with the maximum point of \( |\chi^{(3)}| \). Therefore, when the nonlinear response of the material is at the highest level versus \( \omega_{dc} \) or \( \omega_s \), the frequency estimation of the weak field is minimal while that of the strong field is maximal. Hence, we cannot simultaneously estimate the frequencies
of the driving fields with the best accuracy. Again the HSS is introduced as an efficient figure of merit for revealing the strength of the nonlinear response in the medium.

4. Model II (three-level system)

Now we introduce another model in which a nonlinear medium consisting of three-level atoms is driven by the radiation of amplitude $E_4$ produced in the process of sum-frequency generation. The linear absorption at the frequency of this radiation can be essentially eliminated through the EIT technique. In detail, as illustrated in figure 4, the linear absorption at frequency $\omega_4$ vanishes by an intense saturating field of amplitude $E_s$ at frequency $\omega_s$. Including states $a$, $d$, and $c$ in the atomic wavefunction and working in the interaction picture, we can write the wavefunction as

$$|\psi(t)\rangle = C_a(t)|a\rangle + C_d(t)|d\rangle e^{-i\omega_d t} + C_c(t)|c\rangle e^{-i(\omega_4-\omega_s)t},$$

(13)

satisfying the Schrödinger’s equation (8) with

$$V = -\mu \left( E_4 e^{-i\omega_4 t} + E_s^* e^{i\omega_s t} \right).$$

(14)

Moreover, the Rabi frequencies ($\Omega$, $\Omega_0$) and the detuning factors ($\delta$, $\Delta$) are introduced by the following equations
introduce the linear response of the medium to the laser fields. Again, we see that the qualitative behaviors of the HSS and QFI perfectly resemble. In addition, we computed for the three-level atom, and the linear response of the quantum statistical speeds are

\[ \langle d | V | a \rangle = -\langle d | \mu | a \rangle E_d e^{-i\omega_dt}, \]
\[ \langle e | V | d \rangle = -\langle e | \mu | d \rangle E_e e^{i\omega_et}, \]

\[ \delta \equiv \omega_d - \omega_{da} \quad \text{and} \quad \Delta \equiv \omega_s - \omega_{dc}. \] (16)

Inserting \(|\psi(t)\rangle\) into Schrödinger’s equation (8) and solving the equations of motion for the coefficients \(C \gamma\), one can determine the evolved state of the three-level atom. We intend to solve the differential equations perturbatively in terms of \(\Omega\) but to all orders in \(\Omega\), and take \(C = 0\) for achieving the steady-state solution, leading to [38]

\[ C_a = 1, \quad C_s = -\frac{\Omega - \delta C_d}{\Omega}, \quad C_d = -\frac{\Omega(\delta - \Delta)}{|\Omega|^2 - \delta(\delta - \Delta)}. \] (17)

Calculating the induced dipole moment \(\tilde{p} = \langle \psi | \mu | \psi \rangle = p(\omega_d) e^{-i\omega_dt} + c.c\). and then the polarization \(P = \gamma_{0} \equiv \epsilon_0 \chi^{(1)} E\) leads to the following expression for linear susceptibility [38]:

\[ \chi^{(1)} = \frac{\epsilon_0 \hbar}{N |\mu_{da}|^2} \frac{(\delta - \Delta)}{|\Omega|^2 - (\delta - \Delta)\delta}. \] (18)

The effects of damping can modeled by replacing \(\delta\) by \(\delta + i\gamma_d\) and \(\Delta\) by \(\Delta + i(\gamma_c - \gamma_d)\):

\[ \chi^{(1)} = \frac{\epsilon_0 \hbar}{N |\mu_{da}|^2} \frac{(\delta - \Delta + i\gamma_c)}{(\delta - \Delta + i\gamma_c)}. \] (19)

### 4.1. Relation between strength of linear response and quantum statistical speeds

We aim to explore the relationship among the QFI, HSS, computed for the three-level atom, and the linear response of the medium, composed of a collection of the three-level atoms, to the strong laser field. Again, we see that the qualitative behaviors of the HSS and QFI perfectly resemble. In addition, we introduce the linear response of the medium to the laser fields as a key tool to improve the frequency estimation.

4.1.1. Quantum statistical speeds with respect to \(\omega_{s}\).

Figure 5 shows that when the damping is ignorable, the linear susceptibility may become negative and for definite values of \(\Omega\) or \(\omega\) its sign can be reversed. These definite values may be detected by the QFI and HSS computed with respect to the frequency of the strong laser.

The negative sign of the susceptibility, implying \(n_0 < 1\), can lead to striking interesting phenomena, such as superluminality and parelectricity [83]. The fact that the linear refractive index may be less than unity implies that the phase velocity can be greater than the vacuum speed of light \(c\). Nevertheless, it has been proven that this phenomenon may happen without any violation of special relativity [83]. In fact, the phase velocity, characterizing the velocity of the zero-crossings of the carrier wave, describes the motion of a pattern carrying no information with it [84]. Moreover, the existence of a parelectric medium indicates the possibility of stable electrostatic configurations of charges placed inside an evacuated cavity surrounded by this medium as well as the levitation of an electrical charge in the vacuum above this medium [85].

Comparing \(\chi^{(1)}\) with HSS\(\omega_s(F_{\omega s})\) behavior versus \(\omega_s\), we see when the detuning \(|\delta| = |\omega_d - \omega_{da}|\) in which \(\omega_d = 2\omega + \omega_s\) is high enough, the maximum point of the HSS\(\omega_s(F_{\omega s})\) coincides with the point at which the sign of \(\chi^{(1)}\) is reversed. Otherwise, we see that \(\chi^{(1)}\) and HSS\(\omega_s(F_{\omega s})\) show similar qualitative behavior with respect to \(\Omega\) (see figure 5(a)). Furthermore, comparing their behavior versus \(\omega_s\), we observe that either minimum or maximum point of HSS\(\omega_s(F_{\omega s})\) can detect the sign reversal of \(\chi^{(1)}\) (see figures 5(b) and (c)). Accordingly, the HSS and QFI can be used to detect the passage of the system from subluminality into superluminality.

In the presence of damping, \(\chi^{(1)}\) is not real and its imaginary part, characterizing the linear absorption, can be related to the QFI and HSS. Analyzing the variations of Im\([\chi^{(1)}]\) and HSS\(\omega_s(F_{\omega s})\) versus \(\omega_s\), in figure 6(a), we find that when \(\gamma_{c} \gg \gamma_d\) as well as \(\gamma_c\) is small, and the detuning \(|\delta| = |\omega_d - \omega_{da}|\), in which \(\omega_d = 2\omega + \omega_s\), is high enough, the maximum point of Im\([\chi^{(1)}]\) and HSS\(\omega_s(F_{\omega s})\) coincide. Moreover, under the aforementioned conditions, these measures increase or decrease with each other. Now assume that the detuning \(|\delta| = |\omega_d - \omega_{da}|\) is not high enough. Under these conditions, Im\([\chi^{(1)}]\) and HSS\(\omega_s(F_{\omega s})\) exhibit similar qualitative behavior with respect to \(\Omega\) such that, as shown in figure 6(b), they monotonously decrease with an increase in \(\Omega\). In figure 6(c), we can compare the behavior of Im\([\chi^{(1)}]\) with HSS\(\omega_s(F_{\omega s})\) versus \(\omega_s\). It is found that \(\gamma_{c} \gg \gamma_d\) as well as \(\gamma_c\) is sufficiently small, both Im\([\chi^{(1)}]\) and HSS\(\omega_s(F_{\omega s})\) are minimized for the value of \(\omega_s\) at which the detuning \(|\delta| = |\omega_d - \omega_{da}|\) becomes zero. In addition, they decrease and increase with each other, i.e. there is a harmonious relationship between their variations.

Furthermore, comparing Im\([\chi^{(1)}]\) and HSS\(\omega_s(F_{\omega s})\) behaviors with respect to \(\omega_{dc}\) in figure 6(d), we observe that when \(\delta\)
Figure 5. Three-level system in the absence of damping: (a) normalized quantum Fisher information $F_{\omega}$, Hilbert–Schmidt speed $\text{HSS}_{\omega}$, and linear susceptibility $\chi^{(1)}$ as functions of $\Omega_s$ for $\Omega = 0.00001, \omega_{da} = 20, \omega_{dc} = 1.8, \omega_s = 1.81$; (b) the same quantities versus $\omega$ for $\Omega = 0.001, \Omega_s = 10, \omega_{da} = 20, \omega_{dc} = 2, \omega_s = 2.01$. (c) The same quantities versus $\omega$ for $\Omega = 0.00001, \Omega_s = 100, \omega_{da} = 30, \omega_{dc} = 2.01, \omega_s = 2$.

$= 0$, both measures are minimized for $\omega_{dc} = \omega_s$, leading to $\Delta = \omega_{dc} - \omega_s$ for which the EIT exhibit the best efficiency.

These analyses show that the HSS can be used to monitor the linear absorption of the weak wave in the medium. In addition, this monitoring can be applied to improve the frequency estimation.

4.1.2. Quantum statistical speeds with respect to $\omega$.

In this section, we investigate the usefulness of HSS, computed with respect to $\omega$, for detecting parelectricity, analyzing the absorption experienced by the weak wave in the medium, and improving its frequency estimation.
Figure 6. Three-level system in the presence of damping: (a) Comparison among normalized quantum Fisher information $F_{\omega}$, Hilbert–Schmidt speed $HSS_{\omega}$, and the imaginary part of the linear susceptibility $\text{Im}[\chi^{(1)}]$ versus $\Omega_s$ for $\Omega = 0.00001$, $\omega_{da} = 20$, $\omega = 4.65$, $\omega_{dc} = 1.8$, $\gamma_c = 0.01$, $\gamma_d = 100$; (b) The same quantities versus $\Omega_s$ for $\omega = 9$; (c) the same quantities versus $\omega$ for $\Omega_s = 10$, $\omega_{dc} = 4$, $\gamma_c = 0.001$; (d) the same quantities versus $\omega_{dc}$ for $\Omega_s = 10$, $\omega = 8$, $\omega_s = 4$, $\gamma_c = 0.001$.

Computing the HSS and QFI with respect to $\omega$ we again find that they can detect the sign-reversal of the linear susceptibility. The behavior of $\chi^{(1)}$ and $HSS_{\omega}(F_{\omega})$ versus $\Omega_s$ and $\omega$ is compared in Figure 7 plotted when the damping is ignorable. Figure 7(a) shows that when the detuning $|\delta|$ is high enough, the maximum point of the $HSS_{\omega}(F_{\omega})$ coincides
Figure 7. Three-level system in the absence of damping: (a) normalized quantum Fisher information $F_{\omega}$, Hilbert–Schmidt speed $HSS_{\omega}$, and linear susceptibility $\chi^{(1)}$ as functions of $\Omega_s$ for $\Omega = 0.00001$, $\omega_{da} = 20$, $\omega = 4.65$, $\omega_{dc} = 1.8$, $\omega_s = 1.81$; (b) the same quantities versus $\omega$ for $\Omega = 0.001$, $\Omega_s = 10$, $\omega_{da} = 20$, $\omega_{dc} = 2$, $\omega_s = 2.01$. (c) The same quantities versus $\omega$ for $\Omega = 0.00001$, $\Omega_s = 100$, $\omega_{da} = 210$, $\omega_{dc} = 10$, $\omega_s = 10.01$.

with the point at which $\chi^{(1)}$ experiences the sign reversal, similar to the behavior observed in figure 5(a). Moreover, the sign reversal of $\chi^{(1)}$ can also be detected by searching for the minimum or maximum point of $HSS_{\omega}(F_{\omega})$ versus $\omega$, as shown in figures 7(b) and (c). It should be noted that although maximum and minimum points of $F_{\omega}$ as well as $F_{\omega}$ are witnesses of the sign reversal occurring for $\chi^{(1)}$, we cannot generally conclude that their qualitative behaviors are completely similar, because they may vary oppositely. In other words, for example, when $F_{\omega}$ is maximized (minimized) with respect to $\Omega_s$ or $\omega$, we see that $F_{\omega}$ may be both maximized and minimized (compare figure 5 with figure 7).

According to the aforementioned analyses, we find that not only the HSS and QFI, computed with respect to the frequencies of the driving waves, are efficient tools to detect the passage of the system from subluminality into superluminality, but also monitoring the variations of the linear refractive index can be applied to enhance the frequency estimation.
and HSS\(_{\omega}(F_\omega)\) are minimized for \(\omega_{de} = \omega_\nu\), leading to the best efficiency in the EIT process (see figure 8(c)).

Therefore, in the presence of the damping, there are close relationships among the (HSS, QFI), calculated with respect to the frequency of the driving waves, and the linear absorption experienced by the weak wave.

### 5. Conclusions

In this paper, we investigated how QFI and HSS can be used to characterize the linear and nonlinear responses of an atomic medium to optical fields in two processes: sum-frequency generation and EIT. The atomic medium consists of three and four-level atoms. By computing the QFI and HSS for one of these atoms, we can predict the collective optical behavior of the whole medium. This is one of the main findings of this paper.

We also discovered that the optimal frequency estimation strategy depends on the variations of the (linear and nonlinear) refractive indices of the medium. This has practical implications for measuring nonlinear susceptibilities of optical materials using the Z-scan technique [86–88]. By determining and monitoring the nonlinear refractive index \(n_2\) experimentally, we can achieve the best accuracy in frequency estimation.

Another observation is that the regions at which the linear susceptibility is negative and hence the linear refractive index is smaller than 1, are detectable by the HSS and QFI. Provided that the atomic density \(N\) is sufficiently large, one may expect that in a medium whose linear susceptibility is negative, we have \(\epsilon^{(1)} = 1 + \chi^{(1)} < 0\). Therefore, negative permittivity can also be revealed by the technique presented in this paper. Accordingly, the HSS and QFI are introduced as potential candidates to characterize ferrite materials with negative permeability as well as metamaterials in which permittivity \((\epsilon)\), and magnetic permeability \((\mu)\) are simultaneously negative.

One of the most important motivations for applying nonlinear effects and especially harmonic generation in microscopy is providing enhanced longitudinal and transverse resolution [38]. Nonlinear effects are excited optimally in the region of maximum intensity of a focused laser beam, and hence resolution can be improved. Moreover, the advantage that the signal is far removed in frequency from unwanted background light, resulting from linear scattering of the incident laser beam, is also offered by microscopy based on harmonic generation. Therefore, our results propose the application of HSS and QFI for enhancing image resolutions which is of great importance in various fields of science and technology [76, 89].

Optical damage [90–92] is a practical issue that limits the power and efficiency of nonlinear optical processes by restricting the maximum field strength that can be applied without damaging the optical material. We have demonstrated that the HSS and QFI can determine the optimal Rabi frequency, which is related to the field strength, for exciting the nonlinear response. In this context, the HSS and QFI are proposed to detect optical damage and exceed the damage thresholds in various materials.

**Figure 8.** Three-level system in the presence of damping: (a) comparison among normalized quantum Fisher information \(F_\omega\), Hilbert–Schmidt speed \(\text{HSS}_{\omega}\), and the imaginary part of the linear susceptibility \(\text{Im} [\chi^{(1)}]\) versus \(\omega\) for \(\Omega = 0.00001, \Omega k = 10, \omega_{de} = 20, \omega_{\nu} = 4, \omega_\gamma = 4.001, \gamma_{\nu} = 100, \gamma_d = 0.001\); (b) the same quantities versus \(\omega\) for \(\Omega = 1000, \omega_{de} = 18, \gamma_{\nu} = 100, \gamma_d = 1\); (c) The same quantities versus \(\omega_{de}\) for \(\omega = 8, \omega_\nu = 4, \gamma_{\nu} = 0.02, \gamma_d = 0.01\).
Our work motivates new research on the practical application of the HSS and QFI in designing new materials with artificial magnetism, negative refractive index, and negative refraction, such as super-lenses [37, 75, 93]. Future research could also explore the potential of the HSS and QFI in other areas of physics and engineering.

Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

Acknowledgments

H. R. J. wishes to acknowledge the financial support of the MSRT of Iran and Jahrom University.

Conflict of interest

We have no competing interests.

ORCID iDs

Seyed Mohammad Hosseiny https://orcid.org/0000-0002-5054-9355
Hossein Rangani Jahromi https://orcid.org/0000-0001-7208-2437

References

[1] Helstrom C W 1969 J. Stat. Phys. 1 231–52
[2] Paris M G 2009 Int. J. Quantum Inf. 7 125–37
[3] Giovannetti V, Lloyd S and Maccone L 2006 Phys. Rev. Lett. 96 010401
[4] Giovannetti V, Lloyd S and Maccone L 2011 Nat. Photon. 5 222–9
[5] Degen C L, Reinhard F and Cappellaro P 2017 Rev. Mod. Phys. 89 035002
[6] Pirandola S, Bardhan B R, Gehring T, Weedbrook C and Lloyd S 2018 Nat. Photon. 12 724–33
[7] Pezzè L, Smerzi A, Oberthaler M K, Schmied R and Treutlein P 2018 Rev. Mod. Phys. 90 035005
[8] Huang Z, Macchiavello C and Maccone L 2019 Phys. Rev. A 99 022314
[9] Liu J, Zhang M, Chen H, Wang L and Yuan H 2022 Adv. Quantum Technol. 5 2100080
[10] Rangani Jahromi H and Lo Franco R 2021 Sci. Rep. 11 1–16
[11] Yang J, Pang S, Chen Z, Jordan A N and Del Campo A 2022 Phys. Rev. Lett. 128 160505
[12] Colombo S, Pedrozo-Penafiel E, Adiyatullin A F, Li Z, Mendez E, Shu C and Vuletic V 2022 Nat. Phys. 18 925–30
[13] Garbe L, Bina M, Keller A, Paris M G and Felicetti S 2020 Phys. Rev. Lett. 124 120504
[14] Salvatori G, Mandarino A and Paris M G 2014 Phys. Rev. A 90 022311
[15] Barbieri M 2022 PRX Quantum 3 010202
[16] Fallani A, Rossi M A, Tamascelli D and Genoni M G 2022 PRX Quantum 3 020310
[17] Chu Y and Cai J 2022 Phys. Rev. Lett. 128 200501
[18] Rangani Jahromi H 2019 Int. J. Mod. Phys. D 28 1950162
[19] Petz D and Ghinea C 2011 Introduction to quantum Fisher information Quantum Probability and Related Topics (World Scientific) pp 261–81
[20] Seveso L, Albarelli F, Genoni M G and Paris M G 2019 J. Phys. A: Math. Theor. 53 02LT01
[21] Genoni M G, Olivares S and Paris M G 2011 Phys. Rev. Lett. 106 153603
[22] Zanardi P, Paris M G and Venuti L C 2008 Phys. Rev. A 78 042105
[23] Gacon J, Zoufal C, Carleo G and Woerner S 2021 Quantum 5 567
[24] Šťátník D 2018 Phys. Rev. A 97 042322
[25] Jafarzadeh M, Rangani Jahromi H and Amniat-Talab M 2020 Proc. R. Soc. A 476 20200378
[26] Demkowicz-Dobrzański R, Górecki W and Guţă M 2020 J. Phys. A: Math. Theor. 53 363001
[27] Rangani Jahromi H and Haseli S 2020 Quantum Inf. Comput. 20 0935
[28] Kura N and Ueda M 2020 Phys. Rev. Lett. 124 010507
[29] Veselago V G 1967 Usp. Fiz. Nauk 92 517
[30] Aydın K, Bulu I and Ozbay E 2007 Appl. Phys. Lett. 90 254102
[31] Legrand F, Gérardin B, Bruno F, Laurent J, Lemoulit F, Prada C and Aubry A 2021 Sci. Rep. 11 23901
[32] Fredkin D and Ron A 2002 Appl. Phys. Lett. 81 1753–5
[33] Alù A and Engheta N 2003 IEEE Trans. Antennas Propag. 51 2558–71
[34] Castles F, Fells J A, Isakov D, Morris S M, Watt A A and Grant P S 2020 Adv. Mater. 32 1904863
[35] Zhang Z, Lan Q, Zhang J and Zhou J 2006 J. Mod. Opt. 53 2225–32
[36] Hossain M B, Faruque M R I, Islam S S and Islam M T 2021 Adv. Sci. Rep. 11 1–18
[37] Liu Y and Zhang X 2011 Chem. Soc. Rev. 40 2494–507
[38] Boyd R W 2020 Nonlinear Optics (Academic)
[39] Garmire E 2013 Opt. Express 21 30532–44
[40] Mukamel S 1995 Principles of Nonlinear Optical Spectroscopy (Oxford University Press)
[41] Backus E H, Schafer J and Bonn M 2021 Angew. Chem., Int. Ed. 60 10482–501
[42] Schneider T 2004 Nonlinear Optics in Telecommunications (Springer Science & Business Media)
[43] Cotter D, Manning R, Blow K, Ellis A, Kelly A, Nesson D, Phillips I, Poustit A and Rogers D 1999 Science 286 1523–8
[44] Giezer E, Milosavljevic M, Huang L, Finlay R, Her T-H, Callan J P and Mazur E 1996 Opt. Lett. 21 2023–5
[45] Wu J, Li Z, Luo J and Jen A K–Y 2020 J. Mater. Chem. C 8 15009–26
[46] Leach J, Jack B, Romero J, Jha A K, Yao A M, Franke-Arnold S, Ireland D G, Boyd R W, Barnett S M and Padgett M J 2010 Science 329 662–5
[47] Howell J C, Bennink R S, Bentley S J and Boyd R W 2004 Phys. Rev. Lett. 92 210403
[48] Zhang Z et al 2021 npj Quantum Inf. 7 1–9
[49] Alodjants A, Tsarev D, Ngo T V and Lee R–K 2022 Phys. Rev. A 105 012606
[50] Soileau M, Bass M and Van Stryland E W 1978 Natl. Bur. Stand. Spec. Publ. 541 309–17
[51] Wood R M 2003 Laser-Induced Damage of Optical Materials (CRC Press)
[52] Buscher H T, Tomlinson R G and Damen E K 1965 Phys. Rev. Lett. 15 847
[53] Apostolova T and Ohreshkov B 2022 Appl. Surf. Sci. 572 151354
[54] Weyl G M 2020 Physics of laser-induced breakdown: an update Lasers-Induced Plasmas and Applications (CRC Press) pp 1–67
[55] Braun H H, Döbert S, Wilson I and Wuensch W 2003 Phys. Rev. Lett. 90 224801
