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Raman Scattering of Light and Axions

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Abstract: Based on the modern idea that a photon is a stable, indivisible particle, it is necessary to abandon the generally accepted idea that in the elementary act of Raman scattering of light, a photon splits into two particles. Such a rejection and the transition to a four-photon scheme of the elementary act with the participation of an axion make it possible to preserve the integrity of the photons involved in the elementary act of Raman scattering. The laser beam interacts with vibrations of atoms in molecules, with electrons in atoms, and with phonons or other excitations in the medium under study, as a result of which frequency components appear in the radiation spectrum at the output of the medium under study, shifted relative to the laser frequency toward high or low values. Raman scattering of light is the inelastic scattering of optical radiation in the medium under study, accompanied by a noticeable change in the frequency of radiation. Unlike Rayleigh scattering, which occurs when the refractive indices of the medium and the pumping radiation coincide in phase, in Raman scattering, spectral lines appear in the spectrum of scattered radiation that are not present in the spectrum of primary (exciting) light. Raman scattering of light can be observed in various media: liquids, semiconductors, and gases. It is noteworthy that with the advent of lasers, spontaneous Raman scattering has been supplemented by forced scattering, which allows for a clear result. Using the example of laser radiation scattering in a cell with atomic potassium vapor, it is shown that axions participate in the process of Raman scattering of light.

Keywords: Raman scattering of light, photon, axion, photon annihilation, dispersed medium

1. Introduction

Various regions of the spectrum, spanning from several fractions of an electron volt (eV) to 1 MeV, are searched for a candidate for the role of fundamental particles, the axion (A°). This report analyzes an optical process that fulfills both the necessary and sufficient criteria for the existence of the axion in interactions between electromagnetic radiation and baryonic matter.

The axion decay into two particles $(A°) \to \nu_3 + \nu_{01}$ is the act of this condition, according to theoretical predictions. Based on previously published experimental results, the author proposes that the nearly resonant interaction of optical radiation with atomic, multitier applications in the electric (magnetic) field of an atomic nucleus can isolate these parts of the spectrum, in which the merger pairs photon excitation radiation environment may contribute to the birth of axions: $\nu + \nu \to (A°)$.

The existence of the axion is predicated on the elementary act of two photons resulting from the axion's decay. In earlier optical tests, a portion of the spectrum that is accessible at the output of the photon detection cell with baryonic medium was found in the 1 . . . 2 eV range.

It is in this region of the spectrum that the article discusses examples of such pairs of photons and the reasons why the previously proposed interpretation of the Raman scattering process should not be used.

Later in Section 5 of this paper, issues related to the search for axions in the energy range from a few fractions of an electron volt (eV) to 1 MeV will be discussed.

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According to literature sources, Raman scattering of light is the inelastic scattering of optical radiation by molecules (atoms), accompanied by a noticeable change in the frequency of radiation.

Rayleigh and Raman scattering take place in the quantum environment. In the case of Rayleigh scattering, the frequency of radiation at the outlet of the medium does not change, whereas in the case of Raman scattering, we have scattered radiation components shifted to the Stokes and anti-Stokes spectral regions. If the photons of the pump radiation are absorbed by a two-level quantum system (QM) in an unexcited (ground) state, then its return to the ground state is accompanied by the appearance of Stokes photons in the scattered radiation, whose energy is less than the energy of the primary photons, while part of the energy is transferred to the medium. If the QM in the excited state absorbs the pump radiation's photons, then its return to the ground state is accompanied by the appearance of Raman photons in the scattered radiation, whose energy is greater than the energy of the primary photons due to the transfer of energy from the medium to the photons.

The assumption used in the definition of Raman about metastable levels, to which an electron is transferred in an elementary act, does not explain the mechanism of energy exchange between pump radiation photons and QM electrons. Metastable levels in Raman spectroscopy are associated with the problem of selection rules and the appearance of forbidden lines in the Raman spectra that do not coincide with the absorption lines of the studied QM.

Based on the modern idea that a photon is a fundamental, stable, and indivisible particle, it is necessary to abandon the generally accepted idea that in the elementary act of Raman scattering, a photon splits into two photons (quanta), one of which is absorbed by the medium and the second contributes to Raman scattering.

This rejection, as well as the transition to a four-photon scheme involving an axion, allows for the conservation of the individual identities of the photons involved in the fundamental Raman scattering process. In this case, the axion [1] is a particle born in the electromagnetic field of atomic nuclei due to the annihilation of photon pairs and decays into two new photons, one of which is resonantly absorbed by the medium, due to which the electron passes to the excited level of the atom, and the second photon contributes to Raman scattering.

2. Photons and Axions

Modern science considers the photon as a fundamental elementary particle. In this article, this position is taken as the initial one, which does not require proof. The two-photon Raman scheme, in fact, boils down to the decay of a photon into two parts, one of which is supposedly absorbed by the medium. The abandonment of the two-photon Raman scheme makes it possible to abandon the metastable level and the problem of selection rules, thereby preserving the integrity of the photon.

Efforts to identify a candidate for the elementary particle axion [1, 2] are being carried out across a broad energy spectrum, ranging from fractions of an electron volt (eV) to 1 MeV. This article explores optical processes that fulfill the necessary and sufficient criteria for the existence of an axion in the interaction between electromagnetic radiation and a dispersive medium. Theoretically, this process corresponds to the decay of an axion into two photons occurring in the electromagnetic field of the atomic nuclei of the dispersive medium being studied [2]. Based on the results of experiments performed in atomic vapors of potassium (dispersion medium), Anikin et al. [3] and Ogluzdin [4] posit that, during quasi-resonant interactions of optical radiation with either a two-level system or a multilevel atomic (or other multilevel) medium and within the electromagnetic field of the medium's atomic nuclei, one can identify spectral regions where pairs of photons that excite the medium may fuse (annihilate), thereby contributing to axion generation according to the reaction $h\,\nu + h\,\nu \rightarrow (\mathrm{A}°)$.

A potential confirmation of axion formation and presence could be the appearance of new photon pairs, $(\mathrm{A}°) \rightarrow h\,\nu_3 + h\,\nu_{01}$, which do not originate from the initial excitation source. Here, $h\nu$ is the energy of the pumping photon, $h\nu_3$ is the energy of the Raman photon, and $h\nu_{01}$ is the energy of the photon absorbed by the medium, which coincides with the energy of the inter-level transition of the electron from the ground to the excited level.

Referring to the axion model proposed by Primakoff [2], the generation of two new photons—with energies distinct from those of the pump photons—as a result of axion decay constitutes a necessary and sufficient condition for the axion's existence. This portion of the spectrum is accessible at the output of the potassium vapor cuvette in previous optical experiments by Anikin et al. [3] and Ogluzdin [4].

3. The Need to Introduce an Axion into the Process of Raman Scattering of Light

In Raman scattering, the energy of scattered photons differs from the energy of incident photons – these are photons scattered by the mechanism of Raman scattering. Due to the conservation of energy, the quantum medium either gains or loses energy in the process. As a rule, the energy of incident photons differs from the energy of a quantum transition, but only if the energy of the incident photon is equal to the energy of the quantum transition, such a transition is possible. According to the current theory, the falling quantum disintegrates into parts. What contradicts the definition of a photon is that a photon is a fundamental indivisible particle. In the proposed scheme, the axion decays, which does not contradict its definition. Upon axion decay, the energy of one of the resulting photons matches that of a quantum transition—electronic, vibrational, or rotational—within the medium under investigation. The subsequent nonradiative relaxation facilitates the transfer of this energy to the medium. The second quantum generated from the axion decay exits the medium as a Raman-scattered photon. In this framework, the integrity of all photons involved is maintained.

4. Raman Scattering of Light and Axions

4.1. Interaction between a two-level medium and optical radiation

The following processes are part of the conventional scheme of radiation interaction with a two-level medium, according to Bohr and Einstein: (1) resonant radiation absorption, (2) spontaneous radiation, and (3) resonantly induced radiation. When there is a discrepancy between the radiation source's frequency and the medium's electronic transition frequencies, it is reasonable to believe that a distinct (non-resonant) interaction is responsible for the interaction between radiation and matter. This may be evidenced by the appearance of photons in the scattered radiation, which have a different energy than the photons of radiation at the medium's entrance.

Let's focus on the experiments of Anikin et al. [3] and Ogluzdin [4], the results of which currently suggest that axions are involved in the scattering of photons on potassium atoms when a laser beam passes through a cuvette with atomic vapors of potassium. Recall that, unlike Rayleigh scattering, in the case of Raman scattering, spectral lines appear in the spectrum of scattered radiation that are not present in the spectrum of primary (exciting) light. The number and location of the appearing lines are determined by the structure of the substance.

The experimental setup for studying the interaction of coherent radiation with atomic potassium vapors consisted of (1) a pulse generator on neodymium glass (1.06 mk), (2) a Potassium Dihydrogen Phosphate (KDP) crystal (0.53 mk), (3) a parametric generator on a $LiJO_3$ crystal (0.76 mk), and (4) a cuvette with potassium ($t = 180$–250°C) and a spectrograph.

In Ogluzdin [5], a special case of the interaction of optical radiation with a two-level medium is considered. It is demonstrated that when there is no resonance between the frequency of electronic transitions in the medium and the pump radiation, it is reasonable to suppose that a separate (non-resonant) interaction is responsible for the interaction between matter and radiation. The presence of photons with energy distinct from that of the radiation photons at the medium's entrance in the dispersed radiation may serve as an indicator of this.

For a two-level medium (atomic potassium vapors), it is essential for us that the energy of one of the photons, $h\nu_{01}$, which appeared as a result of decay in the elementary act of axion, coincides with the energy of the transition $4S_{1/2}$-$4P_{1/2}$ (or $4P_{3/2}$), which ensures an electron will resonantly move from the ground level $4S_{1/2}$ to the excited level $4P_{1/2}$ (or $4P_{3/2}$). Once at the excited level of $4P_{1/2}$ (or $4P_{3/2}$), the electron returns to the ground level of $4S_{1/2}$ during nonradiative relaxation, giving energy to the medium (nonradiative relaxation).

The energy of the second photon is equal to:

$$h\,\nu_3 = 2\,h\,\nu - h\,\nu_{01} \qquad (1)$$

where v_{01} is the transition line's frequency between $4S_{1/2}$ and $4P_{3/2}$. The value v_3 in Equation (1) indicates the Raman scattering frequency.

Let's name two reasons that affect the width of the Raman scattering line:

1) Due to the fact that the field strength of the laser beam (unlike the mercury lamp line previously used in Raman spectroscopy) is comparable to the field strength of the atomic nucleus, in the experiment, we have a shift in atomic levels.
2) The field strength of the laser pulse is not constant, due to the changing field strength of the pump radiation during the pulse, the value of the displacement of the level $4P_{1/2}$ changes. Due to the displacement of this level, a change in the magnitude of v_{01} in the ratio (1) (Equation (1)) causes a change in the magnitude of v_3, which in the spectrogram leads to a blurring of the contour of the Raman line.

In the ratio (1) (Equation (1)), the magnitude of 2 $h\,v$ remained the sum of two photons for a long time. The process of transition of this formation into two new photons has not attracted attention for a long time. It was quite natural to turn to the model of annihilation of a pair of photons proposed by Primakoff [2], which states that pairs of photons annihilate to generate an axion in the field of the atomic nucleus, and the latter decays into a new pair of photons—the direct and inverse Primakoff effects. During the decay of the axion, new photons are born, which are absent from the radiation of the pump. Note that in Equation (1), the frequency value of v_3 can be calculated if we know the radiation frequency of the pump source and the frequency of the electronic transition of the medium, which is associated with nonradiative relaxation. The energy value of one of the two photons that appeared after the axion decay is associated with the settlement of this level.

4.2. Interaction of optical radiation with a multilevel medium

We have considered a special case of photon annihilation in an atomic medium. The same processes occur in the molecular environment; as in the case of an atom, we have energy (electronic, vibrational, rotational) levels, with the interaction of pump radiation with QM being either resonant or almost resonant.

Let's consider the process of photon annihilation in the general case for a multilevel QM.

The axion-(A°) can be created when two quanta (photons) in the field of the atomic nucleus are destroyed, according to research by American physicist and scientist Primakoff [2], followed by its decay into a pair of new quanta:

$$h\,v + hv = (A\,^\circ) = hv_{ij} + h\,v_{0j} \tag{2}$$

Where:

$h\,v$ is the energy of the quanta (photons) of the light radiation used to pump QM, with v being the frequency of this radiation.

hv_{ij} represents the energy of quanta (photons) emitted at the exit of the medium, defined by the energy difference between the virtual level-i and the excited electronic, vibrational, or rotational level-j. The element under study has a large number of levels (j) in its QM (atom, molecule) spectrum. We can discuss a range of vibrational, rotational, and electrical levels if the Raman is investigated in a liquid. Therefore, in the general case, at the output of the studied QM, we have a Raman in the form of an expanded spectrum, which includes a set of components v_{ij} [5].

Laser radiation frequency is ν; ν_{01} is the transition line frequency $4S_{1/2}$-$4P_{3/2}$ (13,046 cm^{-1}), and ν_{02} is the transition line frequency $4S_{1/2}$-$4P_{1/2}$ (12,986 cm^{-1}). The Stokes frequency forced electron Raman scattering (FERS) is ν_3. $\nu_3 = 2\nu - \nu_{01}$ (the value of 2ν represents the annihilation of two photons and the production of an axion, during which photons with a frequency of ν_3 occur).

A spectral lamp was used to obtain the reference lines of the doublet ν_{01} and ν_{02} in spectrogram 1b. The radiation frequency of the pump $\nu = 13{,}070$ cm^{-1} is displaced to the anti-Stokes region in the second spectrogram (Figure 1(b)) in relation to the frequency of the transition line $4S_{1/2}$-$4P_{3/2}$. The frequency spectrum is widened in this instance. The spectrogram also shows the faint anti-Stokes FERS line, whose frequency is ν_3*. A shift to the anti-Stokes area of the spectrum with respect to the pumping frequency-ν_{01} implies that axions are also decaying in this region [4].

How does the virtual level's energy get determined? The total energy of the two photons of radiation used to pump QM determines the energy of the virtual level-i; the energy required to move an electron from the ground level to one of the excited levels above j is denoted by v_{0j}.

In Figure 1(a), the Raman expansion of the spectrum in a multilevel system (in our case, a three-level system) is shown by a frequency-limited Stokes wing of the spectrum (ν_s) and a frequency-limited anti-Stokes wing of the spectrum (ν_{as}).

5. Additional Questions

We have considered Raman scattering of light in a dispersion medium, an environment in which the electromagnetic fields of individual atoms are not interconnected and each photon simultaneously interacts with the field of an individual atom.

In Gorelik [6], the characteristics of scalar and pseudoscalar axions in ferroelectrics and amino acids are given. The conditions of photon–pseudoscalara axions energy, which is about 10^{-3} ... 10^{-6} eV, are analyzed. The characteristics of photon–pseudoscalar axion scalar conversion are established. Specific experimental schemes for observing photon-axion conversion in ferroelectrics and amino acids are proposed.

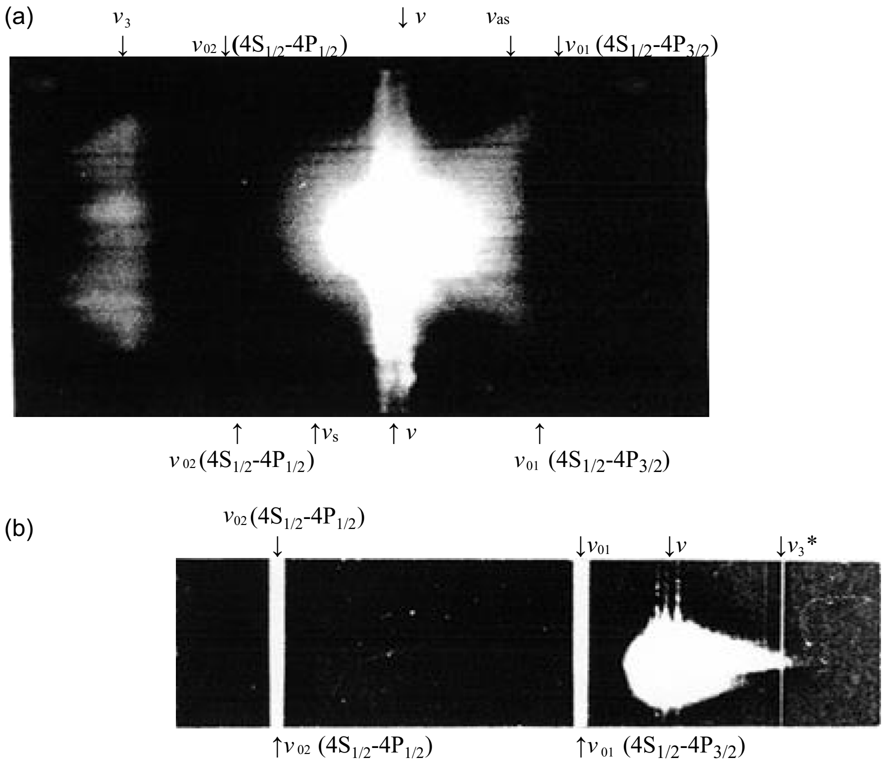
Lin et al. [7] consider the forbidden Raman oscillations of layers (FROL) in layered semiconductor materials (LSM). The intensity distribution of all observed FROL depends on the number of layers, the wavelength of the incident light, and the refractive index mismatch between the LSM and the substrate. These results are explained by the theory of Raman scattering using the proposed spatial interference model, in which natural optical and phonon cavities in the LSM provide spatially coherent photon–phonon coupling mediated by corresponding one-dimensional periodic electronic states. This work shows how the spatial coherence of photonic and phonon fields affects the excitation of phonons using photonic or phonon resonator technology.

Based on the research results, it can be concluded that the Raman spectra of single crystal silicon samples doped with chromium atoms can be used as a tool for assessing their structural perfection and the content of intrinsic and impurity defects, as well as the presence of uncontrolled impurities and oxygen in crystals [8].

In the case of a crystalline medium (KDP crystal), light radiation, propagating in an ordered structure in the absence of absorption, can be converted into a second harmonic [9]. In this case, we have the annihilation of pairs of photons not into axions but into new photons. The difference from the case we are considering is that the frequency of pump radiation in crystals used to produce the second harmonic is in the transparency region

Figure 1

Spectrograms showing how axions contribute to the transmission of laser light pulses in the major doublet frequency range of $4S_{1/2}$ to $4P_{1/2, 3/2}$, via a cuvette containing potassium vapor, (a) pumping radiation frequency $\nu < \nu_{01}$ and (b) pumping radiation frequency $\nu > \nu_{01}$



of these crystals and is far from the absorption band. In addition, in a crystalline medium, the nature of the electromagnetic field does not depend on the arrangement of atoms in a dispersed medium, and the field strength is maintained without changing its magnitude along the entire length of the crystal; in the case of a dispersed medium, the field strength depends on the internuclear distance and is a variable pulsating quantity.

Another case of axion propagation in a crystal structure is considered in Gooth et al. [10]. In this case, axions are particles consisting of Weyl-type electrons (Weyl fermions) in a correlated Weyl semimetal $(TaSe_4)_2I$.

In Kochukov et al. [11], the generation in the Stokes and anti-Stokes spectral regions was observed in a crystal of a cationic solid solution $Sr_{0.9}Ba_{0.1}MoO_4$.

An interesting result for the case of laser beam propagation in a crystal has recently been published in Abrahao et al. [12], in which green laser radiation did not transmit blue laser photons. In this case, it is possible that the radiation of the second harmonic of a non-neodymium laser (0.53 mk) used to illuminate a ruby crystal turns out to be in a situation where photon annihilation leads to the appearance of axions, the decay of which ensures the settlement of the level (0.56 mk) in the ruby crystal. According to Abrahao et al. [12], electrons thrown at this level are able to absorb the radiation of a blue laser propagating in a transverse direction, which creates a shadow on the screen—the radiation of the blue laser does not reach the screen, as in the case of any ordinary physical object with mass. In other words, the authors demonstrate a counterexample—a shadow cast by a laser beam. The observed shadow has the usual characteristics of a typical shadow created by a material object.

Researchers tend to avoid the optical region of the spectrum. Mentioned below are works in which the search for axions is carried out in the energy range from several fractions of an electron volt (eV) to 1 MeV.

The theory of interactions of elementary particles is presented in Belokurov and Shirkov [13].

In Manzari et al. [14], you can find a large list of articles discussing axions and photons in the X-ray range.

In Guendelman [15], the axion–photon system is considered in an external magnetic field in experiments on the mixing of axion photons. The system demonstrates the continuous symmetry of the axion–photon duality in the limit where the mass of the axion can be neglected. The conservation law follows from this symmetry.

Axions can form thermally inside the nuclei of neutron stars, leave the stars due to their weak interaction with matter, and subsequently transform into X-ray radiation in the magnetic fields surrounding the stars. It is shown in Buschmann et al. [16] that the excess of hard X-ray radiation in the energy range of 2–8 keV from isolated neutron stars located nearby can be consistently explained by the existence of an axion-like particle. Currently, there is no generally accepted astrophysical explanation for the excess of hard X-rays.

The paper of Bauer et al. [17] presents new strategies for searching for quadratic interactions of axions and axion-like particles using haloscopes, helioscopes, and quantum sensors, takes into account effects above and below the scale of quantum chromodynamics, and provides a comprehensive sensitivity analysis of current and future experiments.

Hoshino et al. [18] present a comprehensive analysis of the sensitivity of quantum sensors and high-precision measuring instruments to the effects caused by light axions or axion particles. With a mass of less than a few electron volts, axions-light particles can be candidates for the role of dark matter, while the relic density is created due to misalignment, and the field of axions-light particles behaves like a classical (pseudo)scalar background field.

Ning and Safdi [19] are conducting one of the most sensitive searches for ultralight axions to date using data from the Nuclear Spectroscopic Telescope Array (NuSTAR) telescope. They are looking for stellar axions in galaxy M82 with a burst of star formation and in the central galaxy M87 of the Virgo cluster, and then they study their subsequent transformation into hard X-ray radiation in the surrounding magnetic fields. When calculating the luminosity of axions, they take into account all the stellar populations in these galaxies and investigate the conversion of axions into photons using magnetic field profiles in simulated analogues of The Next Generation Illustris Simulation (IllustrisTNG) galaxies. They show that the analysis of NuSTAR data in the range of about 30–70 keV in the direction of these targets did not reveal any signs of axions.

An up-to-date overview of the restrictions can be found in the annually updated handbook by Particle Data Group et al. [20]. Photonic constraints refer to arbitrary axion-like particles, not necessarily the axions of quantum chromodynamics.

6. Conclusion

In classical Raman spectroscopy, the task of qualitative analysis of molecular media was to measure the frequency difference between the fixed frequency of the pump line and the Raman line of the test substance. In the laser era, the demand for Raman spectroscopy has increased.

However, the mechanisms of energy exchange between the medium and pumping photons in the traditional Raman scattering model are still unknown. Referring to the photon annihilation process proposed by Primakov, in our opinion, allowed us to come to an agreement on the issue of photons as fundamental indivisible particles, the four-photon nature of Raman scattering, and the exact resonance that occurs when an electron moves from its ground level to its excited one, which guarantees that energy is transferred to the medium by nonradiative relaxation.

The axion, whose existence was predicted 48 years ago, remains elusive as an elementary particle in extensions of the standard model of particle physics and in the optical frequency domain. Direct registration of the axion in the optical frequency domain is not possible, and we can only indirectly judge its presence from experiments on Raman scattering of light. Studying new properties of axion particles in desktop experiments may allow scientists to better understand the mysterious nature of quantum particles.

Ethical Statement

This study does not contain any studies with human or animal subjects performed by the author.

Conflicts of Interest

The author declares that he has no conflicts of interest to this work.

Data Availability Statement

The data that support the findings of this study are openly available at https://journalofscience.org/index.php/GJSFR/article/view/102768, reference number [5].

Author Contribution Statement

Valeriy Evgenievich Ogluzdin: Conceptualization, Methodology, Software, Validation, Formal analysis, Investigation, Resources, Data curation, Writing – original draft, Writing – review & editing, Visualization, Supervision, Project administration.

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