Do extra compactified dimensions of space cause a substructure underlying the expected eigenstates of a molecule?

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Abstract

Experimental results on NO₂ are explained based on the ADD-model of large extra compactified dimensions of space. We assume that gravity is sufficiently strong in a compactification space of the size of the molecule to affect the vibrational motion of the nuclei by causing an asymmetric perturbation in the symmetric stretch vibrational motion of the optically excited state. At the same excitation energy, there are also other electronic states of different symmetry (conical intersection of potential energy surfaces), which may couple with the optically excited state. Due to the gravitational perturbation the nuclei being in the symmetric stretch vibration mode of the optically excited electronic state pass over into the asymmetric stretch vibration mode of an isoenergetic electronic state. This parity conserving change of the vibronic wave function enhances a small gravitational perturbation to an optically detectable signal. The perturbation is associated with a time constant of about 3 μs, which we attribute to fluctuations of the shape of the compactification space induced by a background cosmic field.

1. Introduction

In this paper we explain experimental results on the triatomic molecule Nitrogen Dioxide (NO₂) based on the theoretical work of Arkani-Hamed, Dimopoulos, and Dvali (ADD) on large extra compactified dimensions of space [1]. In the ADD-model the observed three space dimensions are merely a subspace (a 'brane') in a higher dimensional space, and the three forces of the standard model are confined to this brane but gravity acts in the higher dimensional space [1, 2]. Moreover, gravity may become strong in a compactification space of the size of the molecule. In the present paper we argue that these large extra compactified dimensions affect the quantum states of NO₂ in a way, which is not possible in atoms and diatomic molecules, and we show that unexplained experimental results on NO₂ may well be understood by assuming that the collision-free molecule couples to the hidden extra compactified dimensions of space proposed in the ADD-model.

Triatomic molecules, like NO₂, are the simplest systems in which electronic dynamics is strongly coupled to nuclear dynamics ([3–7] and references given there). For example, laser light (λₜₒₓ = 593 nm) excites NO₂ molecules from the electronic ground state X²A₁ to the excited electronic state A²B₂ [8, 9]. At the same excitation energy, there are also other electronic states (B²B₁ and high lying vibrational levels of the electronic ground state X²A₁). The potential energy surfaces of these electronic states intersect (conical intersection) and enable transitions between these states ([3–7, 10–12] and references given there). For example, the two potential energy surfaces of the electronic states X²Aₐ and A²B₂ intersect at a particular bond angle that depends on the bond length. At this intersection, the nuclei of the molecule being in the symmetric stretch and bending vibration mode in the A²B₂ state may pass over into the asymmetric stretch vibration mode in the X²A₁ state. A small perturbation on the motion of the nuclei enables this parity conserving change of the vibronic wave function (electronic and vibrational motion) of the molecule. The change of the vibronic wave function enhances this perturbation to an optically detectable signal. Thus, the optically excited NO₂ serves as detector of a gravitational
perturbation on the motion of the nuclei in the molecule. The described mechanism is not available in atoms and diatomic molecules, because these systems have none or only one vibration mode, respectively.

Experiments on NO² reveal that underlying an expected eigenstate, a hyperfine structure (hfs) level, of the optically excited NO² is a substructure, which we represent by the two ‘states’ |b⟩ and |c⟩ of the hfs—level. Optical excitation (e.g. near λ_ex = 593 nm or near λ_ex = 514 nm) of the molecule from the ground state |a⟩ populates only |b⟩. Being in |b⟩ the collision-free molecule evolves irreversibly in a non—radiative transition from |b⟩ to |c⟩ in a time τ_b ≈ 3 µs, which is short compared to the radiative lifetime τ_R ≈ 35 µs of both |b⟩ and |c⟩ [13–15], but long compared to the time domain of intramolecular dynamics in NO² (e.g. [16, 17]). The two states |b⟩ and |c⟩ have within the accuracy of the experimental results the same radiative lifetime τ_R but differ in the degree of polarization of the fluorescence light [13, 14]. The transition |b⟩ → |c⟩ preserves the angular momentum couplings, the g-factor, and the Zeeman coherence in the magnetic sublevels. These results imply that the transition |b⟩ → |c⟩ is a change of the vibronic wave function of the molecule. The ‘inversion effect’ (see section 3) shows that light-induced optical coherence between |a⟩ and |b⟩ works against the process causing the transition |b⟩ → |c⟩. Obviously, this process is not caused by electromagnetic interaction. Furthermore, the time constant τ_0 ≈ 3 µs associated with the transition |b⟩ → |c⟩ was measured using different excitation wavelengths, different light sources, and different experimental techniques. We propose associating τ_0 with the action of a background cosmic field, e.g. the axion dark matter field, on the compactification space. The paper is organized as follows. In section 2, the ADD-model is applied to NO², in section 3, we compare this model with experimental results, and section 4 is our conclusion.

2. The ADD-model applied to NO²

The ADD-model was applied to diatomic molecules to study the gravitational force between the nuclei in the molecule due to the extra-dimensional gravitational potential suggested by the ADD-model [18, 19]. The authors analysed the contribution of the gravitational potential to the splitting of vibrational levels in the HD⁺ ion by combining precision measurements and calculations on the HD⁺ ion. An important parameter is the compactification radius R_c of the extra dimensions, which is a measure of the size of the extra—dimensions. Outside the compactification radius, i.e. for radii r > R_c, gravity follows the 3—dimensional Newtonian law, but for r < R_c the extra-dimensional gravitational potential of the ADD—model is effective. Another parameter is the number n_e of extra dimensions. The strength of the gravitational potential depends on both R_c and n_e. The investigations in [19] are setting the upper bound R_c < 0.6 µm on the compactification radius for the case of n_e = 7 extra dimensions. This number of extra dimensions is relevant to M-theory. Based on this result, we apply the ADD-model on the NO² molecule by assuming that gravity is sufficiently strong in a compactification space of the size of the molecule to affect the vibrational motion of the nuclei. Consequently, even the collision-free NO² is not isolated. It is always coupled to the hidden dimensions of space. In the following, we use the expression ‘space-coupled molecule’ as shorthand form for ‘a molecule interacting with the extra compactified dimensions of space’.

We assume that |a⟩, |b⟩, and |c⟩ introduced in section 1 as well as |a'⟩ defined below are states of the ‘space-coupled molecule’, which may be written as

|a⟩ = |a, 1⟩,  |b⟩ = |b, 1⟩,  
|c⟩ = |c, 2⟩,  |a'⟩ = |a, 2⟩  

with ‘1’ and ‘2’ indicating that the ‘space-coupled molecule’ is confined to a single compactification space in |a, 1⟩ and |b, 1⟩ and to two separate compactification spaces in |c, 2⟩ and |a, 2⟩. The state |c, 2⟩ is expected to have a strong asymmetry in the N-O bond length [13, 14]. Obviously in |c, 2⟩ the molecule vibrates in the asymmetric stretch vibration mode having a short and a long N-O bond length with the two nuclei of the short N-O bond length being confined to one compactification space whereas the O nucleus of the long N-O bond length is confined to a separate compactification space. We will further assume that optical transitions conserve the property ‘1’ or ‘2’ of the ‘space-coupled molecule’, because optical transitions have no effect on the extra compactified dimensions of space. Thus, the laser—induced optical transitions between an hfs—level in the ground state and an hfs-level in the excited state of the ‘space-coupled molecule’ are expressed as transitions between the states

|a, 1⟩ ↔ |b, 1⟩  
|a, 2⟩ ↔ |c, 2⟩  

where |a, 2⟩ is no stationary state. The ‘space-coupled molecule’ evolves very fast from |a, 2⟩ to |a, 1⟩ within a few vibrational periods. Therefore, absorption from the ground state starts nearly always from |a, 1⟩ and the transition |a, 2⟩ ↔ |c, 2⟩ is open only for induced emission from |c, 2⟩ to |a, 2⟩.

An essential property of the ‘space-coupled molecule’ is the effect of gravity on the vibrational motion of the nuclei in the molecule. The nuclei of NO² see additionally to the usual potential energy surface (PES) of the
electronic state also the extra-dimensional gravitational potential suggested by ADD-theory (gravitational PES). Both PESs may superimpose symmetrically, i.e. both O nuclei of NO₂ see the same electrical force and the same gravitational force versus the N-O distance. In this case, the gravitational interaction will not change the symmetry type of the vibration mode, but it may eventually introduce anharmonicity in the vibrational motion. However, if the two PESs do not superimpose symmetrically, i.e. for example, if one O nucleus of NO₂ is inside a common compactification space and the other O nucleus is outside this compactification space at the same N-O distance for both O nuclei, we expect that the extra-dimensional gravitational potential is causing an asymmetric perturbation on the symmetric vibrational motion of the molecule, because the gravitational forces on the two O nuclei differ.

In this paper we propose an explanation of the experimental results on NO₂ based on the assumption that the extra-dimensional gravitational potential causes an asymmetric perturbation on the symmetric vibrational motion of the molecule. We attribute the asymmetry to a non-rigid compactification space having a shape, which is fluctuating in time with the characteristic time constant $\tau_0$. In the state $|a,1\rangle$, the molecule is in the totally symmetric ground vibrational level $(0,0,0)$ in the electronic ground state $X^2A_1$. We expect no effect of the extra-dimensional gravitational potential on the symmetry of the vibrational motion of the molecule in $|a,1\rangle$. In the state $|c,2\rangle$, the two nuclei of the short N-O bond length are assumed being confined to one compactification space, and the O nucleus of the longer N-O bond length being confined to a separate compactification space. Also, in this case, we expect no effect of the extra-dimensional gravitational potential on the symmetry of the vibrational motion of the molecule. This is different in $|b,1\rangle$, where the molecule is in the excited symmetric stretch vibration mode in the $A^2B_2$ electronic state and has probably a larger N–O bond length than in the state $|a,1\rangle$ [8, 9]. We assume that the extra-dimensional gravitational potential causes a perturbation on the symmetric vibrational motion of the molecule in $|b,1\rangle$. This perturbation induces randomly asymmetry into the vibrational motion with an average repetition frequency $(1/\tau_0)$.

We introduce a simple model of the perturbed vibrational motion in the state $|b,1\rangle$. The unperturbed symmetric stretch vibration in $|b,1\rangle$ is represented by a sinusoidal oscillation of frequency $\omega_0$ with the autocorrelation function $R_0(T) = (2\pi)^{-1}\cos(\omega_0 T)$ for unlimited duration $T$ of free oscillation. A perturbation affecting the vibrational motion limits the duration $T$ of free oscillation. We assume $R(T) = C \exp(-T/\tau_0) \cos(\omega_0 T)$ as autocorrelation function of the perturbed vibration in $|b,1\rangle$ [20]. Here $C$ is a constant, and $\exp(-T/\tau_0)$ is the probability that the time interval between two perturbations lies between $T$ and $T+\Delta T$. Moreover, we identify the experimentally determined time constant $\tau_0$ with the mean time between two perturbations. $R(T)$ has the power spectrum

$$W(\tilde{\omega} - \tilde{\omega}_0) = \frac{2}{\pi} \int_0^\infty R(T) \cos(\tilde{\omega}T) dT = C \left(\frac{\tau_0}{\pi}\right) \frac{1}{1 + (\tilde{\omega} - \tilde{\omega}_0)^2 (\tau_0)^2}$$

if we consider only positive values of $\tilde{\omega}$ and $\tilde{\omega}_0$. We identify $W(\tilde{\omega} - \tilde{\omega}_0)$ with the probability distribution of vibration frequencies of the perturbed vibrational motion in $|b,1\rangle$. Contrary to the unperturbed symmetric stretch vibration having only $\tilde{\omega}_0$ as vibration frequency, the perturbed vibration comprises a band of frequencies $\tilde{\omega}$ centred at the frequency $\tilde{\omega}_0$. This causes a line broadening of the optical transition $|a,1\rangle \leftrightarrow |b,1\rangle$. We write the resonance frequencies of the optical transition as $\omega_0 = \omega_c + \tilde{\omega}_0$ and $\omega = \omega_c + \tilde{\omega}$, where for instance $\Delta\omega_0$ is the excitation energy of the unperturbed vibrational level of the state $|b,1\rangle$. Therefore, we use $\omega - \omega_0 = (\omega - \omega_c)$ in equation (3) and interpret $W(\omega - \omega_0) = C (\tau_0/\pi) [1 + (\omega - \omega_0)^2 (\tau_0)^2]^{-1}$ as probability distribution of the resonance frequencies of the optical transition $|a,1\rangle \leftrightarrow |b,1\rangle$. The optical resonance frequencies are distributed over a band centred at the frequency $\omega_0$ and having the width (FWHM) $\Delta\omega = 2(\tau_0)^{-1}$ or $\Delta\nu = (\pi\tau_0)^{-1}$. This model predicts an absorption width of the optical excitation of NO₂, which is determined by the time constant $\tau_0$ and not by the radiative lifetime $\tau_R$.

The perturbation causing the line broadening of the optical transition is assumed to induce asymmetry in the symmetric stretch vibrational motion in $|b,1\rangle$, and finally to induce the transition from the symmetric stretch to the asymmetric stretch vibration mode in combination with a change of the electronic state, i.e. the transition $|b,1\rangle \leftrightarrow |c,2\rangle$. However, the perturbation-induced asymmetry in the vibrational motion is in competition with the light-induced optical coherence between $|a,1\rangle$ and $|b,1\rangle$. Optical coherence between $|a,1\rangle$ and $|b,1\rangle$ is keeping the vibrational motion in $|b,1\rangle$ in the symmetry type selected by optical excitation. Therefore, optical coherence works against the transition $|b,1\rangle \leftrightarrow |c,2\rangle$. We will show in section 3 that these two competing processes cause the inversion effect discussed in section 3. The perturbation acting on the vibrational motion in $|b,1\rangle$ is caused by gravitational interaction. It does not affect the optical coherence between $|a,1\rangle$ and $|b,1\rangle$, the angular momentum coupling (rotation, electron spin, nuclear spin), and the Zeeman coherence in the magnetic sublevels in the transition $|b,1\rangle \leftrightarrow |c,2\rangle$.

The model described above is a very rough description of the physical phenomenon that is occurring. The perturbation is caused by the difference $\Delta G$ of the gravitational force acting on the two O nuclei. The model uses an instantaneous perturbation (i.e. a sudden phase change in the vibrational motion). The duration of the
perturbation (the duration with $\Delta G = 0$) is neglected. This approximation seems to be acceptable if the duration of events with $\Delta G = 0$ is short compared to the duration of vibration with $\Delta G = 0$. The extra-dimensional gravitational potential of the compactification space may also induce anharmonicity in the vibrational motion, because the gravitational interaction modifies the potential energy surface of the oscillating nuclei. Anharmonicity affects the vibrational motion of a polyatomic molecule in many ways [21]. However, we expect no significant effect on the model discussed here.

3. Comparison with experimental results

Evidence of the two states $|b, 1\rangle$ and $|c, 2\rangle$ underlying an hfs-level of the optically excited NO$_2$ results from experiments using magnetic field induced depolarization of the fluorescence light (zero-magnetic field level-crossing or Hanle effect) as well as optical radio-frequency double resonance ([13, 14] and references given there). These experiments reveal that the mechanism causing the transition $|b, 1\rangle \rightarrow |c, 2\rangle$ preserves the angular momentum couplings (rotation, electron spin, nuclear spin) and the Zeeman coherence in the magnetic sublevels. We obtain $\tau_B$ and the radiative lifetime $\tau_R$ as coherence decay times e.g. from the width of the ‘broad’ $\langle b, 1\rangle$, $\tau_B$ and the ‘narrow’ $\langle c, 2\rangle$, $\tau_R$ Hanle signal, respectively, with $\tau_R$ being in good agreement with results of radiative decay measurements revealing single-exponential decay ([22] and references given there), and $\tau_B$ being in good agreement with the lifetime $\tau_{in}$ evaluated from measurements of the integrated absorption coefficient giving values for $\tau_{in}$ between 1 $\mu$s and 4 $\mu$s ([23] and references given there). According to theory, $\tau_R$ is the characteristic time of the mechanism that deactivates the upper state $|b\rangle$ of the optical transition [24]. The model in section 2 predicts that $\tau_{in}$ is different from $\tau_R$. This solves an old problem in the spectroscopy of NO$_2$, because $\tau_{in} = \tau_R$ was expected based on the assumption that spontaneous emission is the only mechanism of deactivation of the upper state of the optical transition. The discrepancy of $\tau_{in}$ and $\tau_R$ by more than a factor 10 stimulated extensive work on the ‘anomalously’ long radiative lifetime [25, 26]. On the other hand, the disagreement of $\tau_{in}$ with $\tau_R$ and the result $\tau_{in} \approx \tau_R$ is a strong support for the model of the two states $|b, 1\rangle$ and $|c, 2\rangle$ and the non-radiative transition $|b, 1\rangle \rightarrow |c, 2\rangle$ due to the asymmetric perturbation acting on the symmetric vibration in $|b, 1\rangle$.

An unusual experimental result associated with the transition $|b, 1\rangle \rightarrow |c, 2\rangle$ is the inversion effect, which is not seen on atoms and diatomic molecules [13, 14, 27, 28]. The important parameters in this effect are the light intensity I of the exciting laser light and the transit time $T_t$ of the molecules in a molecular beam through the interaction length with the exciting laser light. The effect is seen in the Hanle signal and in the optical radio-frequency double resonance signal associated with the state $|c, 2\rangle$ as a change from a downward to an upward pointing resonance signal versus I or $T_t$ [13, 14, 27]. The width of both signals is independent of I and $T_t$. A phenomenological description of this effect assumes that optical coherence between $|a, 1\rangle$ and $|b, 1\rangle$ reduces the transition rate $|b, 1\rangle \rightarrow |c, 2\rangle$ [14, 28]. This agrees with the model in section 2. The model predicts two competing processes in $|b, 1\rangle$. One process is the perturbation inducing asymmetry in the vibrational motion in $|b, 1\rangle$, which causes finally the transition $|b, 1\rangle \rightarrow |c, 2\rangle$. The other process is the ‘oscillation’ of the molecule between $|a, 1\rangle$ and $|b, 1\rangle$ due to the light-induced optical coherence between $|a, 1\rangle$ and $|b, 1\rangle$. The initial absorption from $|a, 1\rangle$ to $|b, 1\rangle$ is followed by a subsequent stimulated emission from $|b, 1\rangle$ back to $|a, 1\rangle$ and another absorption from $|a, 1\rangle$ to $|b, 1\rangle$, and so on. This process keeps the vibrational motion in $|b, 1\rangle$ in the symmetry type selected by optical excitation. For low light intensity the probability for induced emission is too small to reduce the flow of population from $|b, 1\rangle$ to $|c, 2\rangle$. However, with increasing light intensity, optical coherence works against the transition $|b, 1\rangle \rightarrow |c, 2\rangle$ and reduces the flow of population from $|b, 1\rangle$ to $|c, 2\rangle$.

The following example makes the inversion effect plausible. We consider the optical transition $|a, 1\rangle \leftrightarrow |b, 1\rangle$ with $\Delta m = 0$ excitation of the magnetic sublevels $|b, 1, m\rangle$ of $|b, 1\rangle$. For small values of I and $T_t$, the $|b, 1, m\rangle$ and the $|c, 2, m\rangle$ have the same occupation probability distribution, because the transition $|b, 1\rangle \rightarrow |c, 2\rangle$ is independent of the quantum number m. Those magnetic sublevels in $|b, 1\rangle$ and $|c, 2\rangle$ have maximum occupation probability, for which the optical transition $|a, 1\rangle \leftrightarrow |b, 1\rangle$ has maximum transition probability. For large values of I and $T_t$, the occupation probability distribution in the $|b, 1, m\rangle$ has still its maximum at the same sublevels as for small values of I and $T_t$, but the occupation probability distribution in the $|c, 2, m\rangle$ is inverted. The magnetic sublevels $|c, 2, m\rangle$, which have maximum occupation probability for small values of I and $T_t$, have minimum occupation probability for large values of I and $T_t$, because the flow of population from $|b, 1, m\rangle$ to $|c, 2, m\rangle$ has a minimum for these magnetic sublevels. To our opinion the inversion effect reveals clearly that the perturbation inducing asymmetry in the symmetric vibration in $|b, 1\rangle$ is a gravitational interaction, because an electromagnetic interaction would easily affect the optical coherence between $|a, 1\rangle$ and $|b, 1\rangle$.

The flow of population from $|b, 1\rangle$ to $|c, 2\rangle$ determines mainly the absorption properties of NO$_2$, because the radiative decay of $|b, 1\rangle$ is by a factor of $\tau_0/\tau_R \leq 0.1$ smaller than the transition rate $|b, 1\rangle \rightarrow |c, 2\rangle$. On the other hand, this transition rate depends on the light-induced optical coherence between $|a, 1\rangle$ and $|b, 1\rangle$ as the
inversion effect shows. Perturbations on the light-induced optical coherence between \(|a, 1\rangle\) and \(|b, 1\rangle\) e.g. by a static magnetic field or a radio-frequency field (rf-field) cause an increase in the absorption of \(NO_2\) or equivalently an increase in the intensity \(F_{\text{tot}}\) of the fluorescence light. We investigated two types of resonances in the intensity of the fluorescence light, the ‘broad rf- resonance’ \([29]\) and the ‘\(\sigma\) — resonance’ \([30]\). The width of these resonances depends on the time constants \(\tau_0\) and on the transit time \(T_1\), but not on the light intensity \(I\). For vanishing transit time broadening \((1/T_1 \to 0)\), one obtains \(\tau_0 \approx 3 \mu s\) from both resonance signals. As was shown in particular for the ‘broad rf- resonance’, these resonance signals are strongly related with the inversion effect \([28]\). Using these resonances in the intensity of the fluorescence light, we measured \(\tau_0\) on several absorption lines in the 593 nm band with use of a single-mode cw dye laser and near 514 nm with use of a tuneable single-mode Ar-ion laser \([30]\). The experimental arrangements for the ‘broad rf- resonance’ and the ‘\(\sigma\) — resonance’ are the same as in the Hanle and optical radio-frequency double resonance experiments, which both represent a resonant change in the polarization of the fluorescence light. The superposition of the resonances in the intensity of the fluorescence light with the resonances in the polarization of the fluorescence light was not recognized e.g. in the early Hanle experiments \([30—32]\). Reproducible measurements on the ‘\(\sigma\) — resonance’ and the ‘broad rf- resonance’ were obtained after we used a reproducible adjustment procedure of the transit time \(T_1\) \([27,33]\).

The model in section 2 is strongly supported by the experiments reported in \([33]\), where two light beams \(L_1\) and \(L_2\) intersect a beam of freely propagating \(NO_2\) molecules with an adjustable spatial separation between \(L_1\) and \(L_2\), defining a time of flight of the molecules between \(L_1\) and \(L_2\) up to about the lifetime \(\tau_0\) of the molecules. The light of a single mode laser is split into the two beams \(L_1\) and \(L_2\) with \(L_1\) interacting first with the molecules and inducing the optical transition \(|a, 1\rangle \leftrightarrow |b, 1\rangle\), and with \(L_2\) interacting with the molecules in \(|a, 1\rangle\) and in \(|c, 2\rangle\). These experiments show that the \(NO_2\) molecules travel collision-free in the molecular beam during a time of flight of up to 35 \(\mu s\). The absorption width (FWHM) of the laser-induced transition \(|a, 1\rangle \leftrightarrow |b, 1\rangle\) is \(\Delta \nu = (\pi \tau_0)^{-1}\) in good agreement with the model in section 2. There is no direct absorption from \(|a, 1\rangle\) to \(|c, 2\rangle\).

The state \(|c, 2\rangle\) is populated via the optical transition \(|a, 1\rangle \leftrightarrow |b, 1\rangle\) and the subsequent radiation-less transition \(|b, 1\rangle \to |c, 2\rangle\). \(L_2\) interacts with the molecules in \(|c, 2\rangle\) by induced emission from \(|c, 2\rangle\) to \(|a, 2\rangle\) in agreement with the model in section 2.

4. Discussion and conclusion

In this paper, we show that unexpected experimental results on the molecule \(NO_2\) are well described by the gravitational coupling of the vibrational motion of the nuclei in the molecule to the large extra compactified dimensions of space suggested by Arkani-Hamed, Dimopoulos, and Dvali (the ADD-model \([11]\)). The experiments on \(NO_2\) reveal a substructure underlying an expected eigenstate, a hyperfine structure (hfs)-level, of the optically excited \(NO_2\). We describe this substructure by the two states \(|b, 1\rangle\) and \(|c, 2\rangle\) of the hfs-level. The molecule is excited into \(|b, 1\rangle\) by an optical transition from the ground state \(|a, 1\rangle\). The extra-dimensional gravitational potential suggested by the ADD-model induces asymmetry in the symmetric stretch vibration mode in \(|b, 1\rangle\) and causes the molecule to evolve irreversibly from \(|b, 1\rangle\) to \(|c, 2\rangle\) in a time \(\tau_0 \approx 3 \mu s\). In \(|c, 2\rangle\) the molecule vibrates in the asymmetric stretch vibration mode. The transition \(|b, 1\rangle \to |c, 2\rangle\) is possible due to the ‘conical intersection’ (see section 1) in the optically excited state of the molecule. The transition \(|b, 1\rangle \to |c, 2\rangle\) results in several optical resonance signals and in a broadening of the vibrational levels, which determines the absorption properties of this molecule. The time constant \(\tau_0\) is the same on all investigated absorption lines in \(NO_2\), and we conclude that \(\tau_0\) is not a property of the molecule.

We assume a compactification space having about the size of the molecule in the ground state \(|a, 1\rangle\) and in the excited state \(|b, 1\rangle\). The N–O bond length in \(NO_2\) is about 1.3 \(x 10^{-10}\) \(\mu m\) in \(|b, 1\rangle\) and slightly smaller in \(|a, 1\rangle\) \([8]\). However, the model proposed in the present paper assumes that in the state \(|c, 2\rangle\) the molecule \(NO_2\) extends over two compactification spaces. The state \(|c, 2\rangle\) is assumed to have a short and a long N–O bond length with the two nuclei of the short N–O bond length being confined to one compactification space whereas the O nucleus of the long N–O bond length is confined to a separate compactification space. The values of the bond lengths in \(|c, 2\rangle\) are not known. However, a bond length exceeding \(10^{-3}\) \(\mu m\) is very unlikely as calculations e.g. in \([12]\) show. The extension of \(NO_2\) over two compactification spaces in the state \(|c, 2\rangle\) determines also the allowed optical transitions according to equations \((2a)\) and \((2b)\). The restrictions imposed by these equations are necessary for a consistent description of the experimental results. Therefore, it is necessary to assume an extension of \(NO_2\) over two compactification spaces in the state \(|c, 2\rangle\). This is setting the upper bound \(R_c < 10^{-3}\) \(\mu m\) on the compactification radius \(R_c\). This result lowers the upper bound for the compactification radius \(R_c\) considerably compared to the value \(R_c < 0.6 \mu m\) reported in \([19]\). But contrary to the discussion in \([19]\), the model proposed in the present paper does not specify the number \(n_e\) of the extra dimensions. As \(R_c\) depends on the number \(n_e\) of the extra dimensions, it may be possible to evaluate \(n_e\) from the result \(R_c < 10^{-3}\) \(\mu m\). However, this is beyond the
scope of the work reported here. Finally, we note also that a scenario, in which each atom of NO$_2$ is in a separate compacification space, is not able to describe the experimental results. This is setting a lower bound on the compacification radius $R_c$.

We propose a non-rigid compacification space having a shape, which is changing in time with the characteristic time constant $\tau_0$. However, this does not necessarily mean a change of the volume of the compacification space. The change of the shape of the compacification space causes an asymmetric gravitational perturbation on the symmetric stretch vibration in the state $|b, 1\rangle$, because the nuclei of the molecule see a time-dependent asymmetry of the extra-dimensional gravitational potential as discussed in section 2. This is an addendum to the ADD-theory, because the experimental results are not in agreement with a rigid compacification space. We attribute the change of the shape of the compacification space with the time constant $\tau_0$ to the action of a background cosmic field. A justification of this proposal is beyond the scope of the present work reported here and needs further clarification by theory. A possible candidate for the cosmic field is the axion dark matter field, which is expected to act as a coherently oscillating classical scalar field in the kHz—MHz frequency band ([34, 35] and references given there). The time constant $\tau_0 \approx 3 \mu s$ fits well to this frequency band. If we identify the decay rate $(1/\tau_0)$ with the oscillation frequency of the axion field we obtain $m c^2 \approx 200$ peV for the mass $m$ of the axion [34, 35].

If $\tau_0 \approx 3 \mu s$ is a property of the space, we expect to find this time constant also in other molecules for instance in the lifetime $\tau_{in}$ obtained from the integrated absorption coefficient. The molecules NO$_2$, SO$_2$, and CS$_2$ represent a class of small polyatomic molecules whose radiative lifetimes $\tau_0$ are more than one order of magnitude longer than the lifetimes $\tau_{in}$ obtained from their integrated absorption coefficients [25]. The present study shows that the gravitational perturbation on the vibrational motion broadens the vibrational levels of NO$_2$ and determines the absorption properties of NO$_2$. The disagreement of $\tau_{in}$ with $\tau_0$ and the result $\tau_{in} \approx \tau_0$ is a strong support for the model considered here. Unfortunately, the measured values for $\tau_{in}$ vary strongly. For instance, first measurements of $\tau_{in}$ in NO$_2$ resulted in $\tau_{in} \approx 0.3 \mu s$ [36]. Later experiments showed that $\tau_{in} \approx 3 \mu s$ is a more realistic value [23]. Lifetimes $\tau_{in}$ are also reported for CS$_2$ ($\tau_{in} \approx 3 \mu s$) and for SO$_2$ ($\tau_{in} \approx 0.2 \mu s$) [25]. Possibly, molecular spectroscopy on small polyatomic molecules offers a new way for a search into physics beyond the Standard Model.

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