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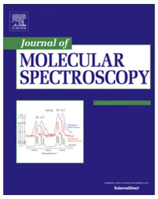

Photoassociation of ultracold molecules near a Feshbach resonance as a probe of the electron–proton mass ratio variation

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ABSTRACT

We show that the photoassociation (PA) rate of ultracold diatomic molecules in the vicinity of a Feshbach resonance could be used to probe variations of the electron-to-proton mass ratio $\beta = m_e/m_p$, a quantity related to other fundamental constants via the $\Lambda_{\rm QCD}$ scale. The PA rate exhibits two features near a Feshbach resonance, a minimum and a maximum, which are very sensitive to the details of the interactions and the exact mass of the system. The effect and detection threshold are illustrated in the formation rates of ultracold Li_2 and LiNa molecules by performing coupled-channel calculations in an external magnetic field. We find that the PA rate is particularly sensitive near narrow Feshbach resonances in heavy molecules, where it might be possible to detect relative variability of β on the order of 10^{-15}—10^{-16}. We also use a two-channel model to derive a proportionality relation between the variation of the PA rate and β applicable to diatomic molecules.

1. Introduction

Quasar absorption spectra hint that the fundamental constants may have changed over the history of the Universe [1–8]. If proved correct, there would be a profound impact on the fundamental laws of physics. Perhaps most importantly, the equivalence principle in general relativity would be violated, leading to a change of our understanding of cosmology [9]. Modern theories aiming to unify the four fundamental forces often allow temporal and spatial variations of the fine structure constant $\alpha = e^2/\hbar c$ and the electron–proton mass ratio $\beta = m_e/m_p$ due to the dramatic changes in the Universe's structure during its evolution. Grand unification theories also suggest more than one mechanism to make all coupling constants and elementary particle masses time- and space-dependent. A detailed review of the connection between the fundamental constants and their significance in different theoretical frameworks can be found in Ref. [10].

In addition to astrophysical observations, limits on detection of the space–time variability of fundamental constants have been established from the Oklo natural nuclear reactor [11,12], Big Bang Nucleosynthesis [13], cosmic microwave background [14], meteorites [15], and precision laboratory measurements using optical and microwave atomic fountain clocks [16–19]. A good summary of the present day values is given in Refs. [2,3,20,21]. So far these experiments have not detected a variation of any of the fundamental constants. However, this neither confirms nor contradicts the quasar spectra observations that probe very different epochs in the evolution of the Universe.

Recent developments in ultracold atomic and molecular physics offer new possibilities for precision measurement experiments [22]. On one hand, atomic and molecular spectra depend on α through relativistic corrections which are observable as the fine structure, Lamb shift, hyperfine structure, and other small perturbations. On the other hand, molecular bound levels and certain types of molecular couplings depend on the mass, making phenomena depending on them (such as Feshbach resonances in ultracold atomic samples [23]) particularly well adapted to investigate the relative variation of dimensionless electron–proton mass ratio $\delta\beta/\beta$ over time. This can be understood as molecules are bound by the electronic interaction (making molecular interaction potential electronic in origin), while the mass of the individual atoms is mostly baryonic. Consequently, the scattering and spectroscopic properties of ultracold molecules, such as the scattering length or the binding energy of the last vibrational energy level, are very sensitive to β, and particularly suitable atomic and molecular systems have quasi-degenerate levels caused by the coupling terms of different nature. Presently, the most accurate model-free laboratory measurement limits the magnitude of relative temporal variation of β to less than about 4×10^{-14}/year, based on a comparison

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of the frequency of a rovibrational transition in SF_6 with the fundamental hyperfine transition in Cs [24]. A number of different approaches to measuring the variation of β in atomic and molecular systems has been proposed [22,25–32]. The suggested experiments may be able to detect a time variation on the order of $\dot{\beta}/\beta \sim 10^{-15}$/year or better [10,26,33], independently of the results obtained by the atomic clocks.

In one suggested approach, Chin and Flambaum have studied the variation of β near a Feshbach resonance and derived a proportionality relation between the relative variations of the zero-energy atom–atom scattering length [23] and electron–proton mass ratio. The proportionality factor between the two quantities can be very large for suitable systems, implying that an indirect method for detecting small changes in β could be based on measuring the relative variation of the scattering length near a Feshbach resonance. This technique can be applied to any diatomic system as long as Feshbach resonances can be found or induced.

Motivated by the result of Ref. [23] and growing interest in a search for variability of fundamental constants, we propose a new approach to accurately measure temporal variation of β using photoassociative Feshbach spectroscopy in an ultracold gas. Photoassociative spectroscopy is an accurate and robust technique that has been used successfully to obtain high resolution spectra of molecular states in optically cooled gases [34,35], such as two-photon spectrocopy [36]. It also allowed studies of the scattering length between atoms [37–39], and of the formation of ultracold molecules in their ground electronic states [40–42]. It has been predicted theoretically [43–45] and observed in experiments [46] that the PA rate of diatomic molecules near a magnetic Feshbach resonance can be enhanced by several orders of magnitude. The enhancement is caused by a large increase in the short-range amplitude of the open-channel component of the scattering wave function, leading to higher probability density and increased dipole transition rates between the scattering state and a target excited state at small internuclear separation.

In this article, we demonstrate the enhanced sensitivity of two features in the PA rate (a minimum and a maximum) near a Feshbach resonance to the variability of β, should such variation exist in Nature. Our calculations suggest that an even larger proportionality factor than predicted in Ref. [23] can be expected between these quantities in suitable diatomic systems. We investigate in more detail the enhancement factor at the PA minimum, which is particularly interesting for potential experimental realizations as its position does not depend on the laser intensity [47,48], nor suffers from the perturbations present in the strongly interacting regime associated with the very large scattering length at the resonance.

2. Theory

The PA rate coefficient K_b, for a molecule photoassociated in a bound ro-vibrational level $b = (v,j)$, where v and j stand for the vibrational and rotational quantum number, respectively, of a target electronic state from a pair of colliding atoms, can be expressed as $K_b = \langle v_{\rm rel}\sigma_b\rangle$, where $v_{\rm rel}$ is the relative velocity of the approaching atoms and σ_b is the PA cross section for the corresponding bound level b. Here, the brackets indicate averaging over the relative velocity distribution of the colliding atoms. If a Maxwell–Boltzman distribution of velocities characterized by the temperature T is assumed, the PA rate coefficient becomes [49,50].

$$K_b(T) = \frac{k_BT}{hQ_T}\int_0^\infty |S_b(\varepsilon,\ell,\omega)|^2 e^{-\varepsilon/k_BT}\,\frac{d\varepsilon}{k_BT}, \qquad (1)$$

where $Q_T = (2\pi\mu k_BT/h^2)^{3/2}$ is the translational partition function and $\varepsilon = \hbar^2k^2/2\mu$ is the asymptotic kinetic energy for the colliding atom pair of reduced mass $\mu, \hbar\omega$ is the photon energy, k_B is Boltzmann constant, $S_b(\varepsilon,\ell,\omega)$ is the S-matrix element for the PA process, and ℓ is the quantum number describing the relative angular momentum of the colliding atoms. Near an isolated bound level b, the S-matrix has the form [49]

$$|S_b(\varepsilon,\ell,\omega)|^2 = \frac{\gamma_b\gamma_s}{(\varepsilon-\Delta_b)^2 + \frac{1}{4}(\gamma_s+\gamma_b)^2}, \qquad (2)$$

where $\Delta_b = E_b - \hbar\omega$ is the photon frequency detuning relative to the ro-vibrational level $b = (v,j)$ of the target molecular state of energy E_b, γ_b is its natural linewidth, and $\gamma_s(\varepsilon,\ell)$ is the stimulated linewidth resulting from the laser coupling. Here, all other possible decay processes, such as predissociation and ionization, are neglected. We note that in the unitarity limit, where $|S|^2 = 1$, the maximum value of the PA rate coefficient (1) is given by

$$K_b^{\max} = \frac{k_BT}{hQ_T} = \frac{h^2}{(2\pi\mu)^{3/2}}\frac{1}{\sqrt{k_BT}}. \qquad (3)$$

For low PA laser intensities, where multi-photon or ionization processes can be neglected, γ_s can be obtained using the Fermi's golden rule, resulting in [49,50]

$$\gamma_s(\varepsilon,\ell) = \frac{\pi I}{\epsilon_0 c}|\langle b|D(R)|\varepsilon,\ell\rangle|^2, \qquad (4)$$

where ϵ_0 and c are the vacuum permittivity and speed of light, respectively, I is the PA laser intensity, $|\varepsilon,\ell\rangle$ and $|b\rangle$ are the energy-normalized initial continuum state and the final bound ro-vibrational state, respectively, $D(R)$ is the transition dipole moment between these states, and R is the internuclear separation. Both the initial and final state wave functions depend on the internuclear separation.

If an external magnetic field is applied, it breaks the degeneracy of molecular hyperfine states and produces a Zeeman shift of the energy levels of the interacting atoms. A magnetic Feshbach resonance occurs if the energy of a bound ro-vibrational level in the molecular ground state in closed channel is shifted by the field to coincide with the asymptotic energy of the colliding atoms [51].

In the presence of a single magnetic Feshbach resonance, the real part of the atom–atom scattering length a can be parametrized as a function of the magnetic field B according to [52]

$$a(B) = a_{\rm bg}(B)\left(1 + \frac{\Delta B}{B - B_0}\right), \qquad (5)$$

where ΔB is related to the width of the Feshbach resonance (proportional to the coupling strength between the scattering channels), and B_0 is the resonant magnetic field. The background scattering length $a_{\rm bg}(B)$ depends on the magnetic field weakly and can be assumed to be constant. A typical behavior of the scattering length $a(B)$ is illustrated in Fig. 1(a) for a s-wave Feshbach resonance in ^{7}Li (see next section for details).

Photoassociation in an external magnetic field can be described by a two-channel model that includes one open channel $|{\rm O}\rangle$ with amplitude $\Psi_{\rm O}(R)$ and a closed channel $|{\rm C}\rangle$ with amplitude $\Psi_{\rm C}(R)$, typically coupled by hyperfine and Zeeman interactions (Fig. 2) [43]. The total s-wave ($\ell = 0$) scattering state can then be written as

$$|\Phi\rangle_{\rm tot} = \Psi_{\rm O}(R)|{\rm O}\rangle + \Psi_{\rm C}(R)|{\rm C}\rangle, \qquad (6)$$

with

$$\Psi_{\rm O}(R) = \psi_{\rm reg}(R) + \tan\delta\,\psi_{\rm irr}(R) \qquad (7)$$

$$\Psi_{\rm C}(R) = -\sqrt{\frac{2}{\pi\Gamma}}\sin\delta\,\psi_0(R), \qquad (8)$$

where $\psi_{\rm reg}$ is the "regular" scattering wave function, *i.e.* without coupling to the closed channel, and $\psi_{\rm irr}$ is the "irregular" scattering wave function arising from that coupling. The amount of coupling is

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