Coefficients for sensitivity of fine-structure transitions in carbonlike ions to α variation

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We calculate sensitivity coefficients to α variation for the fine-structure transitions (1,0) and (2,1) within $^3P_J[2s^22p^2]$ multiplet of the carbonlike ions C I, N II, O III, Na VI, Mg VII, and Si IX. These transitions lie in the far-infrared region and are in principle observable in astrophysics for high redshifts $z\sim 10$. This makes them very promising candidates for the search for possible α variation in a cosmological time scale. In such studies one of the most dangerous sources of systematic errors is associated with isotope shifts. We calculate isotope shifts with the help of relativistic mass shift operator and show that it may be significant for C I but rapidly decreases along the isoelectronic sequence and becomes very small for Mg VII and Si IX.

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I. INTRODUCTION

Some of the extensions of the standard model predict small space-time variations of such fundamental constants as the fine-structure constant $\alpha=e^2/(\hbar c)$ and electron-to-proton mass ratio $\mu=m_e/m_p$. These variations are now intensively searched for both in astrophysical data and in laboratory experiments. For this purpose, two or more lines with different dependencies on the fundamental constants are compared to each other at different times. Laboratory experiments provide extremely high sensitivity to frequency shifts, while the typical time scale is of the order of one year. The astrophysical extragalactic observations allow studying time intervals comparable to the lifetime of the universe ($\sim 10^{10}$ yr), but with much lower accuracy. Different theoretical models predict different behavior of the constants from monotonic in time, to oscillations and sharp changes. The latter could take place, for example, during the transition from matter-dominated to dark-energy-dominated universe.

We can conclude that astrophysical and laboratory methods are complementary and equally important. Recent progress and perspectives of the laboratory searches were summarized in [1]. At present all laboratory experiments give only strong upper bounds on the time variation in fundamental constants. Situation in astrophysics is less clear. Some of the observations indicate nonzero variation at a few-sigma level [2–5], while other results are consistent with zero variation [6–8]. For more references and the discussion of the most recent developments see online material from the workshop [9].

Controversial astrophysical results may indicate some systematic effects. One of the possible systematic frequency shifts can arise from the different velocity distributions of different species in molecular clouds, i.e., the so-called Doppler noise [10,11]. Another potentially dangerous systematic effect may be associated with the cosmological evolution of the isotope abundances, which can lead to the time-dependent isotope shifts of atomic transitions [2,12]. The former effect can be suppressed when different lines of the same specie are used to study possible variation in constants. The latter systematics is absent in microwave spectra of molecules, where lines of different isotopes are well resolved. Alternatively, one can use either atoms with single stable isotope, or specific combinations of atomic frequencies, which are not sensitive to isotope shifts [13]. In order to apply this latter method one needs to know accurate values of the isotope shift coefficients.

An interesting opportunity to search for α variation is associated with M1 transitions between fine-structure (FS) levels of ground multiplets. These lines are observable for very large cosmological redshifts up to $z\sim 10$ [14,15], and have high sensitivity to α variation. In the case of ^2P_J multiplet of C II ion there is only one FS line (3/2,1/2). As a reference one can use microwave molecular lines, which are insensitive to α variation, but depend on the mass ratio μ. This way one can put a limit on the variation in the combination $F=\alpha^2/\mu$ [11]: $\Delta F/F=(0.1\pm 1.0)\times 10^{-4}$ at $z=6.42$ (the look back time about 13 Gyr). The signal in this method is enhanced by the large and different sensitivities of compared lines. On the other hand, the lines of different species are used, so the Doppler noise can become a problem when the accuracy is increased.

Many atoms and ions have triplet ground state ^3P_J, and two M1 transitions (1,0) and (2,1) can be observed. The most important species of this kind for astrophysical observations are carbonlike and oxygenlike ions. In this case one can measure the ratio of these two frequencies and compare it to the laboratory value. This way the Doppler noise can be significantly suppressed. However, in the first approximation within the LS coupling scheme, the FS frequencies obey the Landé rule,

$$\omega_{2,1}/\omega_{1,0}=2. \quad (1)$$

Thus, the frequency ratio does not depend on α.

Sensitivity coefficients $\mathcal{Q}$ to α variation are defined as

$$\Delta\omega/\omega = 2\mathcal{Q}\Delta\alpha/\alpha. \quad (2)$$

In approximation (1) these coefficients are the same for both FS transitions,

$$\mathcal{Q}_{2,1} = \mathcal{Q}_{1,0} = 1. \quad (3)$$

Equations (1) and (3) break when we take into account nondiagonal spin-orbit interaction. Equation (1) also breaks when Breit interaction is taken into account. If the latter is neglected, one can link experimentally observed violation of the Landé rule with the difference in sensitivity coefficients [16],

$$\Delta\mathcal{Q} = \mathcal{Q}_{2,1} - \mathcal{Q}_{1,0} = \frac{1}{2}\left(\frac{\omega_{2,1}}{\omega_{1,0}}\right) - 1. \quad (4)$$

This simple expression predicts significant differences in sensitivity coefficient of FS transitions, which rapidly grow with nuclear charge Z. In fact, Breit interaction cannot be neglected for the light ions with $Z \lesssim 10$. For this case, Eq. (4) significantly overestimates $\Delta\mathcal{Q}$.

For the configuration ns^2np^2 one can take Breit interaction into account within well-known semiempirical theory [17]. Corresponding results for a number of ions of astrophysical interest are given in [16]. However, this theory is essentially one configurational and does not account for certain correlation corrections. The latter can be adequately treated only within relativistic *ab initio* calculations.

In this paper we report *ab initio* calculations of the sensitivity coefficients $\mathcal{Q}$ for FS transitions in the ground multiplets of carbon and carbonlike nitrogen, oxygen, sodium, magnesium, and silicon. Our results are in a good agreement with analytical estimates [16], the differences being typically on the order of 10%. All mentioned elements, with exception of sodium, have several stable isotopes. Inhomogeneity of isotope distribution and cosmological evolution of isotope abundances can lead to the isotope shifts of observed lines in comparison to the laboratory frequencies. This can cause systematic effects for α variation studies. Unfortunately, the isotope shifts for the FS transitions are not known. For this reason we calculated isotope mass shift coefficients k_{ms} for all ions with several isotopes. For the ions considered here the volume shift is much smaller and can be neglected.

Among the light elements C, N, and O (CNO), it is mainly carbon where the cosmic evolution may change isotopic ratios considerably. The ^{12}C:^{13}C ratio is about 90 in the solar system, but it can be as low as 6 in the atmospheres of evolved massive red supergiants such as Alpha Ori [18]. The enhanced ^{13}C abundance relative to ^{12}C is caused by the CNO cycle operating in massive stars. Therefore, one can expect that in high redshift objects, where only short lived, massive stars can have evolved, the ^{12}C:^{13}C ratio can be much below the solar value and a ^{13}C fraction of up to 20% is possible in the early universe. The question of ^{12}C:^{13}C evolution has also been discussed by Fenner *et al.* [19] for somewhat later phases where intermediate mass stars convert ^{12}C into ^{13}C in the asymptotic giant branch. Here peak values of ^{13}C:^{12}C of 0.06 are predicted. For nitrogen, the ^{14}N:^{15}N ratio is always large so that isotopic shifts are probably not important. However, in the Murchison meteorite, the ^{14}N:^{15}N ratio varies in SiC grains between 10^2 and 4×10^3, and the lower values of ^{14}N:^{15}N have been found in grains with low ^{12}C:^{13}C [20]. For oxygen, the solar system value ^{16}O:^{17}O is about 2630, but this ratio can be lower in CNO processed material by a factor of 5 (Alpha Ori [18]). This means that there may be some cosmic evolution of isotopic ratios in N and O, however at a much lower level than in C, and not detectable at the present achievable spectral resolution. For the evolution of the isotopic ratios of Mg and Si, which have both several abundant isotopes, we refer to the works of Ashenfelter *et al.* [21] and Fenner *et al.* [19]

II. DETAILS OF THE CALCULATIONS

We used Dirac-Breit Hamiltonian and all-electron configuration-interaction (CI) method. It was shown in Ref. [16] that deviation from the Landé rule was caused primarily by nondiagonal spin-orbit interaction between the levels $^3P_{0,2}$ and $^1S_0$, $^1D_2$, while the level $^3P_1$ remained practically unperturbed. Therefore, it was essential that the theory accurately reproduced the spacings for all five levels of the ground configuration. Below we present our results for all levels of interest and compare theoretical energies with the experimental data from NIST [22].

Most calculations were done with Dirac-Sturmian basis set [23,24], which included orbitals up to $9s$, $9p$, $8d$, $8f$, and $7g$. Configurational space corresponded to triple and partly quadruple excitations from $1s$, $2s$, and $2p$ shells. We also did calculations with a much smaller configurational space which included singles for $1s$ shell and doubles for $2s$ and $2p$ shells, but on a somewhat longer basis set. In this smaller calculation we neglected retardation part of the Breit interaction. We found out that retardation corrections were very small. Expansion of the configurational space was more important and led to better agreement with the experiment for the transition frequencies. On the other hand, the difference for the sensitivity coefficients was not dramatic (see below). Correlation effects were most important for neutral carbon and less important for the carbonlike ions.

Comparison of the results for frequencies and $\mathcal{Q}$ factors from the calculations with two configurational spaces described above confirmed that the smaller space was sufficiently saturated. That allowed us to use it for the calculations of the isotope shifts and significantly reduce the computational costs.

In order to determine $\mathcal{Q}$ factors we calculated atomic energy levels for three or five values of α in the vicinity of its physical value and performed numerical differentiation. Three point differentiation was used in most previous calculations [25,26]. Here we were interested in the small differences in $\mathcal{Q}$ factors for the FS levels, so we compared three-point differentiation with more expensive five-point differentiation. The difference appeared to be rather small.

Calculations of the mass shift coefficients are similar to calculations of $\mathcal{Q}$ factors [27,28]. The Hamiltonian H_{ms}, which describes mass shift, is added to atomic Hamiltonian with the coefficient λ: $H_\lambda = H + \lambda H_{\text{ms}}$. After solving eigenvalue problem for H_λ one finds the mass shift coefficient by numerical differentiation, $k_{\text{ms}} = \partial E_\lambda / \partial\lambda$.

TABLE I. Energies in cm^{-1} and $\mathcal{Q}$ factors for the levels of the configuration $2s^22p^2$ with respect to the ground state $^3P_0$. Experimental frequencies are taken from Ref. [22].

	$^3P_1$			$^3P_2$			$^1D_2$			$^1S_0$		
Ion	Expt.	Theor.	$\mathcal{Q}$	Expt.	Theor.	$\mathcal{Q}$	Expt.	Theor.	$\mathcal{Q}$	Expt.	Theor.	$\mathcal{Q}$
C I	16.42	16.4	0.9998	43.41	43.3	0.9948	10192.6	10435	0.0023	21648.0	21879	0.0014
N II	48.7	48.3	1.0086	130.8	129.3	1.0004	15316.2	15605	0.0052	32688.8	33036	0.0031
O III	113.2	112.8	1.0197	306.2	304.9	1.0040	20273.3	20535	0.0099	43185.7	43540	0.0058
Na VI	698	693	1.0671	1859	1850	1.0132	35498	35719	0.0375	74414	74795	0.0215
Mg VII	1107	1108	1.0891	2924	2920	1.0172	40948	41192	0.0534	85153	85571	0.0306
Si IX	2545	2532	1.1403	6414	6394	1.0254	52926	53185	0.0990	107799	108253	0.0573

For the mass shift we use relativistic theory developed in [29–32]. For optical transitions in light atoms the relativistic corrections to mass shift are usually small [33]. However, here we are interested in the infrared FS transitions. As long as the fine-structure disappears in the nonrelativistic limit, the nonrelativistic theory of the isotope shift is not applicable here.

In the nonrelativistic approximation the total mass shift is a sum of the normal mass shift (NMS) and the specific mass shift (SMS). The NMS contribution comes from the substitution of the electron mass with the reduced mass, and respective coefficient is simply proportional to the transition frequency: $k_{\mathrm{nms}}^{\mathrm{nr}}=\omega/1823$ a.u. The SMS contribution is described by the two-electron operator $\Sigma_{i\neq k}\boldsymbol{p}_i\cdot\boldsymbol{p}_k$ and has to be calculated numerically. In the relativistic theory the NMS and SMS contributions correspond to one-electron $(i=k)$ and two-electron $(i\neq k)$ parts of the relativistic mass shift operator,

$$H_{\mathrm{MS}}=\sum_{i,k}\left(\boldsymbol{p}-\frac{\alpha Z}{2r}[\boldsymbol{\alpha}+(\boldsymbol{\alpha}\cdot\hat{\boldsymbol{r}})\hat{\boldsymbol{r}}]\right)_i\cdot\left(\boldsymbol{p}-\frac{\alpha Z}{2r}[\boldsymbol{\alpha}+(\boldsymbol{\alpha}\cdot\hat{\boldsymbol{r}})\hat{\boldsymbol{r}}]\right)_k, \tag{5}$$

which is correct to the second order in αZ. Here NMS contributions have to be calculated numerically on the same footing as SMS [29–33].

III. NUMERICAL RESULTS AND DISCUSSION

Results of our calculations of the energies and sensitivity coefficients $\mathcal{Q}$ for the levels of the ground configuration $1s^22s^22p^2$ are presented in Table I. All of them are given with respect to the ground state $^3P_0$. Calculations are done for the "large" configurational space described in Sec. II. This space includes $\sim 1.2\times 10^5$ relativistic configurations.

We see that for the levels of the ground multiplet the $\mathcal{Q}$ factors are close to unity, as expected. For two other levels of the ground configuration the $\mathcal{Q}$ factors are small. Deviation from unity in the former case and deviation from zero in the latter are of the same order of magnitude and rapidly grow with nuclear charge Z. Here we are mostly interested in the differences of the $\mathcal{Q}$ factors for the FS transitions (1-0) and (2-1). These differences are given in Table II.

From Table II one can see that our numerical results are in good agreement with semiempirical estimates from Ref. [16]. Calculations with "large" and "small" CI described above are very close to each other. Except for the neutral carbon, the difference between two numerical calculations is much smaller than their difference from semiempirical values. Note that for the large CI we used five-point differentiation method and three-point method for the small CI. Therefore, we can conclude that small CI is already sufficiently saturated and that three-point differentiation is also sufficiently accurate. Because of that, in the following calcula-

TABLE II. The differences of the sensitivity coefficients $\Delta\mathcal{Q}$ of the FS emission lines within the ground multiplet ^3P_J for the most abundant carbonlike ions. *Ab initio* results of this work are compared with semiempirical results [16] based on the theory [17]. In addition to the large CI we also made a much smaller CI as described in the text.

	Transition a (1,0)		Transition b (2,1)			$\Delta\mathcal{Q}=\mathcal{Q}_b-\mathcal{Q}_a$		
Ion	λ_a (μm)	ω_a (cm^{-1})	λ_b (μm)	ω_b (cm^{-1})	ω_b/ω_a	Ref. [16]	Large CI	Small CI
C I	609.1	16.40	370.4	27.00	1.646	−0.008	−0.0081	−0.0090
N II	205.3	48.70	121.8	82.10	1.686	−0.016	−0.0132	−0.0134
O III	88.4	113.18	51.8	193.00	1.705	−0.027	−0.0250	−0.0247
Na VI	14.3	698	8.6	1161	1.663	−0.091	−0.0861	−0.0843
Mg VII	9.0	1107	5.5	1817	1.641	−0.12	−0.116	−0.114
Si IX	3.9	2545.0	2.6	3869	1.520	−0.21	−0.190	−0.188

TABLE III. Coefficients $k_{\rm nms}$ and $k_{\rm sms}$ for carbonlike ions. Mass shift is calculated with respect to the ground state $^3P_0$. All numbers are in units of GHz amu. For NMS we also give nonrelativistic value $k^{\rm nr}_{\rm nms}=\Delta E_{\rm exper}$(in units of a.u.)$\times 3609.6$.

		$^3P_1$	$^3P_2$	$^1D_2$	$^1S_0$
C I	$k_{\rm nms}$	−0.09	−0.42	178.8	369.7
	$k^{\rm nr}_{\rm nms}$	0.27	0.71	167.6	356.0
	$k_{\rm sms}$	0.51	1.75	−155.2	−151.2
N II	$k_{\rm nms}$	−0.31	−1.26	263.5	553.2
	$k^{\rm nr}_{\rm nms}$	0.80	2.15	251.9	537.6
	$k_{\rm sms}$	1.35	4.42	−179.4	−167.4
O III	$k_{\rm nms}$	−0.09	−1.43	343.3	725.2
	$k^{\rm nr}_{\rm nms}$	1.86	5.04	333.4	710.2
	$k_{\rm sms}$	2.56	8.79	−197.0	−165.6
Mg VII	$k_{\rm nms}$	7.33	14.51	661.9	1398.3
	$k^{\rm nr}_{\rm nms}$	18.21	48.09	673.4	1400.4
	$k_{\rm sms}$	21.58	62.86	−221.5	131.1
Si IX	$k_{\rm nms}$	19.83	43.96	834.0	1748.3
	$k^{\rm nr}_{\rm nms}$	41.86	105.49	870.4	1772.9
	$k_{\rm sms}$	45.34	124.24	−197.6	468.3

tions of the mass shifts we use this significantly cheaper method (small CI includes ~10 000 relativistic configurations).

Our results for the mass shifts are summarized in Tables III and IV. Table III gives coefficients $k_{\rm nms}$ and $k_{\rm sms}$, while Table IV presents full mass shifts for given isotope pairs. We see that for the FS transitions within the ground state the NMS coefficients calculated with the help of relativistic operator [Eq. (5)] differ very strongly from the nonrelativistic values. Interestingly, the difference is bigger for the light ions, where even the sign of the nonrelativistic approximation is wrong. Relativistic NMS coefficient becomes positive between $Z=8$ (oxygen) and $Z=9$ (fluorine) but remains significantly smaller than nonrelativistic predictions. Note that

TABLE IV. Frequency shifts $\delta\nu=\nu^{A'}-\nu^{A}$ for FS transitions $(J'-J)$ in isotopes A' and A of C-like ions. The last two columns give respective velocity shifts $\delta V=-(\delta\nu/\nu)c$, where c is the speed of light.

			$\delta\nu$ (GHz)		δV (km/s)	
Ion	A'	A	(0-1)	(1-2)	(0-1)	(1-2)
C I	13	12	−0.00278	−0.00602	1.691	2.228
N II	15	14	−0.00498	−0.01006	1.024	1.225
O III	17	16	−0.00909	−0.01798	0.803	0.932
	18	16	−0.01717	−0.03396	1.517	1.760
Mg VII	25	24	−0.04819	−0.08076	0.435	0.444
	26	24	−0.09267	−0.15531	0.837	0.855
Si IX	29	28	−0.08026	−0.12689	0.315	0.328
	30	28	−0.15517	−0.24532	0.610	0.634

for two optical transitions between levels of different multiplets the relativistic and nonrelativistic values are very close. This result confirms that nonrelativistic theory of the mass shift works nicely for optical transitions but is not applicable to FS transitions.

Table III shows that SMS coefficients also significantly change along the isoelectronic sequence. The NMS coefficients are mostly sensitive to relativistic effects, which depend on the parameter αZ. The SMS coefficients are more sensitive to correlations. The latter are governed by the parameter $1/Z$ and decrease along the sequence. It is clearly seen from Table IV that full mass shift for the FS transitions behaves more smoothly than NMS and SMS parts. This may mean that in the relativistic theory there is no good reason to separate mass shift into NMS and SMS parts. The last two columns of Table IV give apparent velocity (Doppler) shifts in astrophysical observations, which correspond to the frequency shifts from two previous columns. We see that velocity shifts for two FS transitions significantly differ for light ions, but become almost equal for heavier ions. For Mg VII and Si IX the isotope shifts practically cancel out from the frequency ratio $\omega_{2,1}/\omega_{1,0}$. Consequently, for these ions the isotope shift does not lead to noticeable systematic effect in α-variations studies.

IV. CONCLUSIONS

FS transitions can be observed in far-infrared waveband for very distant astrophysical objects with redshifts $z\sim 10$. This makes them very promising candidates for the study of possible α variation on the cosmological time scale. We performed *ab initio* calculation of the sensitivity coefficients $\mathcal{Q}$ to α variation for the FS transitions within ground multiplet ^3P_J of carbonlike light ions. These calculations confirmed that the differences in sensitivity for FS transitions $\Delta\mathcal{Q}$ are of the same order as $\Delta\mathcal{Q}$ for optical transitions in the same ion. In both cases $\Delta\mathcal{Q}$ rapidly grows with Z. In optical waveband the most dangerous systematic effect is associated with the isotope shifts. We calculated mass shifts for FS transitions and found that isotope shift is rather large for light ions C I, N II, and O III, but practically cancels out for Mg VII and Si IX. At the same time these heavier ions, together with the Na VI ion, which has only one stable isotope, have higher sensitivity to α variation.

It was pointed out in Sec. I that in the solar system light elements C, N, and O have only one dominant isotope. However, in the early universe the abundances of ^{12}C and ^{13}C could be significantly different. At the same time, the isotope shift for the FS transitions in C I is larger than for other C-like ions, while sensitivity to α variation is lower. We conclude that the method suggested in [16] may not work for C I because of the small sensitivity and large systematic effects associated with isotope shifts. For C-like ions the sensitivity to α variation grows with the nuclear charge Z, while isotope shifts rapidly decrease. Moreover, for heavier ions the isotope shifts almost cancel out for the ratio of the FS frequencies. Another dangerous systematic effect from the Doppler noise is significantly suppressed for the pairs of lines of the same species. Therefore, observations of the FS transitions in heavier C-like ions can be used as a sensitive tool for the search of α variation at large redshifts.

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