

angle of resolution of the detector, $f_0 \leq 1$. In this case we should point out that in an experiment carried out on silicon crystals,¹¹ the TDS intensity may amount to up to 40% of the overall scattering intensity in the Bragg reflection. Judging from the estimate given above, we can only expect this fraction to increase for protein crystals.

TDS may significantly influence the determination of $\langle x^2 \rangle$, if $f_0 \sim 1$. In fact, $\langle x^2 \rangle$ is extracted from the experimental data by determining the dependence of $\log P$ on Q^2 . From Eqs. (9)–(11) we obtain

$$\log P = -(1-f_0)\langle x^2 \rangle Q^2 + \text{const}, \quad (12)$$

if $Q^2 \langle x^2 \rangle \ll 1$ (for the first Bragg peak, $Q^2 \langle x^2 \rangle \sim 10^{-3}$), so that the experimental slope proves to be $-(1-f_0)\langle x^2 \rangle$, and not $-\langle x^2 \rangle$.

Thus, for a crystal comprised of large biomolecules, f_0 may be ~ 1 ; and consequently the experimentally determined $\langle x^2 \rangle$ in x-ray diffraction will be significantly less than the true value.

In the case of RSMR, TDS is easily separated from the elastic scattering by the Mössbauer detector, and consequently the determination of $\langle x^2 \rangle$ by this technique should be more accurate.

CONCLUSION

Thus, in our work we determined the amplitude of the mean square atomic displacement $\langle x^2 \rangle$ for myoglobin in the crystalline state by the technique of measuring the Rayleigh scattering of the Mössbauer radiation (RSMR). We have taken the RSMR spectra and determined the fraction of elastic scattering for crystalline myoglobin, the buffer, glycerine, and water. We developed a tech-

nique to separate $\langle x^2 \rangle$ for the Mb molecules from a crystalline sample comprised of molecules of buffer solution and myoglobin. The value of $\langle x^2 \rangle$ experimentally measured using RSMR proved to be significantly greater than the $\langle x^2 \rangle$ measured by x-ray diffraction. We show that in the case of crystals comprised of large biomolecules, thermal diffusive scattering (TDS) strongly affects the value of $\langle x^2 \rangle$ experimentally determined from x-ray diffraction, leading to a significant under-estimation of these values compared with the true ones determined by RSMR.

In conclusion, the authors express their thanks to V. A. Namiot for useful discussions.

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¹Ravnesnaya dinamika nativnoy struktury belka (Equilibrium Dynamics of the Native Protein Structure), Pushchino (1977).

²H. Frauenfelder, G. A. Petsko, and D. Tsernoglou, *Nature* **280**, 558 (1979).

³P. J. Artymiuk, C. C. F. Blake, D. E. P. Grace, S. J. Oatley, D. C. Phillips, and M. J. E. Sternberg, *Nature* **280**, 563 (1979).

⁴D. C. Champeney and G. W. D. Woodhams, *J. Phys. B* **1**, 620 (1968).

⁵D. C. Champeney and G. W. Dean, *J. Phys. C* **8**, 1276 (1975).

⁶E. É. Gaubman, Yu. F. Krupyanskiy, V. I. Gol'danskiy, T. V. Zhuravleva, and I. P. Suzdalev, *Zh. Éksp. Teor. Fiz.* **72**, 2172 (1977). [*Soviet Physics JETP* **45**, 1141 (1977)].

⁷F. Parak and H. Formanek, *Acta Cryst., Sect. A* **27**, 573 (1971).

⁸International Table for X-Ray Crystallography, Kynoch, London (1962).

⁹D. C. Champeney and D. F. Sedgwick, *J. Phys. C* **5**, 1903 (1972).

¹⁰C. Kittel, *Introduction to Solid State Physics*, Wiley, New York, 1971. (Russ. Transl., Nauka, 1978, p. 76).

¹¹G. Albanese, C. Ghezzi, A. Merlini, and S. Pace, *Phys. Rev. B* **5**, 1746 (1972).

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Nonresonant many-photon ionization of atoms in a strong stochastic field

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The probability of nonresonant ionization is calculated for the case of atoms in a strong stochastic low-frequency electromagnetic field. The calculations are made with accuracy up to the coefficient of an exponential function. It is found that, as in the case of a monochromatic field, the probability of ionization by a stochastic field is determined by a single parameter; it is called the stochastic adiabatic parameter. The well-known limiting cases of a stepwise many-photon process and of tunnel ionization are discussed. A general expression is derived for the statistical factor which characterizes the ratio of the ionization probabilities in stochastic and monochromatic fields with the same radiation intensity. The known experimental results agree well with the calculations.

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The study of the features of the process of ionization of atoms by a field of stochastic radiation is a subject of present importance in theoretical and experimental spectroscopy. After the fundamental work of Keldysh,¹ who investigated the basic laws of the ionization of

quantum systems by a strong coherent electromagnetic field, the development of powerful lasers made it possible to study this process experimentally for specific atomic systems (see, for example, Refs. 2 and 3 and the literature they cite). However, since present-

day lasers produce radiation which, strictly speaking, is incoherent, definite difficulties are encountered in comparing the experimental data with the theory. Besides this, it is often better to use for experiments laser radiation with a large number of longitudinal modes. Both theoretical⁴⁻⁷ and experimental⁸ analyses show that the intensity distribution of such radiation is close to an exponential law; i.e., the statistics of this radiation is Gaussian.

Accordingly, the qualitative agreement of the experimental results on many-photon ionization of atoms with the theory calls for additional theoretical investigation of this process, with the stochastic character of the laser radiation taken into account. Some features of the ionization of quantum systems by stochastic radiation have been given in several papers.⁹⁻¹¹ In particular, specific numerical calculations have been done of the so-called statistical factor G , which is the ratio of the probabilities (W and w) of ionization of an atom by fields of stochastic (W) and of monochromatic (w) radiation with the same value of the field-strength amplitude. These calculations were done for five-photon ionization of xenon atoms^{10,11} by radiation from a neodymium laser. These results, however, are not of a sufficiently universal character.

A more general theoretical analysis of the effect of the coherence of radiation on the process of many-photon ionization of an atom was made in a paper by Karapetyan.⁹ On the basis of that work we make in this paper a numerical universal calculation of the statistical factor for nonresonant ionization of atoms, using the adiabatic approximation and working to exponential accuracy.

The most interesting region for experiments is that of low frequencies ($\omega \ll E_0$, where E_0 is the energy of the discrete ionization level) and high intensities \mathcal{E} of the field. One example is the ionization of inert gas atoms observed in fields with intensities 10^6 – 10^8 V/cm.¹²⁻¹⁴ Under these conditions the probability of ionization by a monochromatic field with frequency ω and intensity \mathcal{E} is given (to exponential accuracy) by¹

$$w = \exp[-2nf(\gamma)], \quad (1)$$

$$f(\gamma) = \left(1 + \frac{1}{2\gamma^2}\right) \operatorname{arsh} \gamma - \frac{1}{2\gamma}(1 + \gamma^2)^{1/2}, \quad (2)$$

where $n = E_0/\omega$, i.e., n is equal to the multiplicity of the many-photon ionization. The condition for the adiabatic approximation is $n \gg 1$. The quantity

$$\gamma = \omega(2E_0)^{1/2}/\mathcal{E}$$

is called the adiabatic parameter. A system of units is taken in which $m = e = \hbar = 1$.

Accordingly, many-photon ionization by a monochromatic field is characterized by a universal function of a single parameter γ . The form of this function is shown in Fig. 1. For $\gamma \ll 1$ we have from Eq. (2)

$$f(\gamma) = 2\gamma/3$$

and then from Eq. (1) we get the probability of the tunnel transition

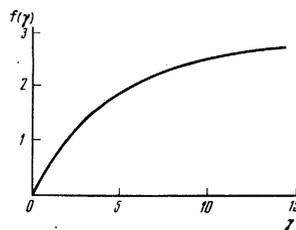


FIG. 1. The universal function $f(\gamma)$ of the adiabatic parameter γ , which determines the probability of many-photon ionization in a monochromatic field.

$$w = \exp[-4(2E_0)^{3/2}/3\mathcal{E}], \quad (3)$$

which agrees, up to a multiplicative constant, with the probability of ionization by a constant electric field. In the opposite limiting case $\gamma \gg 1$ we find from Eq. (2) that $f(\gamma) = \ln(2\gamma/e^{1/2})$, so that from Eq. (1) we get

$$w = (e^{1/2}\mathcal{E}/2\omega(2E_0)^{1/2})^{2n}. \quad (4)$$

This is nothing other than the result of n -th order perturbation theory. A power law for the ionization probability is realized at not too high values of the field strength \mathcal{E} , of the order of 10^4 – 10^6 V/cm,¹²⁻¹⁵ as we see from the definition of the adiabatic parameter γ .

In a stochastic (Gaussian) field the ionization probability is given by the expression

$$W = \frac{2}{\mathcal{E}^2} \int_0^\infty \exp\left\{-2nf(\gamma) - \frac{\mathcal{E}^2}{\mathcal{E}^2}\right\} \mathcal{E} d\mathcal{E}, \quad (5)$$

where $\mathcal{E} = (\overline{\mathcal{E}^2})^{1/2}$ is the rms value of the intensity of the stochastic field. Our problem is to calculate the expression (5) with exponential accuracy.

Before doing the calculation, let us consider the question of when the averaging procedure defined by Eq. (5) is justified. In principle one should determine the amplitude for the ionization transition in the non-monochromatic field and then take the square of its absolute value. Instead of this we take the square of the absolute value of the amplitude as calculated for a monochromatic field, and then average it over the various harmonics. These harmonics correspond to the Fourier series expansion of an external field with magnitude \mathcal{E} varying randomly with time.

This approach to the averaging of the probability is obviously justified when the field strength amplitude \mathcal{E} varies randomly and sufficiently slowly. From the mathematical point of view this means neglecting the damping of the correlation function $\langle \mathcal{E}(t)\mathcal{E}(0) \rangle$ of the field. This is justified when the characteristic time τ of the random variations of the field amplitude is large in comparison with the transition time $1/\omega$. This approximation, which is valid far from thresholds of many-photon transitions, can be violated near such thresholds, i.e., when with change in frequency the minimum number of photons necessary for ionization changes by a unit. In fact, when this is so the quantity w is small and the condition $\tau \gg 1/\omega$ may not be satisfied. The thresholds are clearly marked only in weak fields, or more precisely under the condition $\gamma \gg 1$,

when the ionization is many-photon in nature (see later discussion). In such a case allowance for the damping of the correlation function would lead to a smearing out of the ionization thresholds. In the tunnel-transition regime ($\gamma \ll 1$) and in the intermediate case ($\gamma \sim 1$) the condition $\tau \gg 1/\omega$ is always satisfied for realistic strengths \mathcal{E} of a laser electromagnetic field, which obviously must not be supposed too small.

Since we are considering the case $n \gg 1$, we apply the method of steepest descent to calculate the integral (5). The result is

$$W = \exp[-2nF(\bar{\gamma})], \quad (6)$$

where the quantity F is determined as the solution of the transcendental equation

$$-F + \frac{1}{2} \operatorname{sh} 2F = \bar{\gamma}. \quad (7)$$

The quantity $\bar{\gamma}$ that appears in Eq. (7) is given by

$$\bar{\gamma} = \omega(2\omega)^{1/2} / \mathcal{E}$$

and can be called the stochastic adiabatic parameter.

Accordingly, in a stochastic field the probability of an ionization process is also determined as a universal function of a single parameter, $F(\bar{\gamma})$. This function is shown in Fig. 2. There are analytic limiting expressions for it. For $\bar{\gamma} \ll 1$ we find

$$F(\bar{\gamma}) = (3/2 \bar{\gamma}^2)^{1/2}$$

and for the probability W we get the well known expression⁹

$$W = \exp[-2 \cdot 3^{1/2} E_0 / \mathcal{E}^{3/2}]. \quad (8)$$

Like Eq. (3), the expression (8) is the probability of a tunnelling. We see that in a stochastic field, as in a monochromatic field, the dependence on the field frequency ω disappears, but there is a change in the dependence on \mathcal{E} and E_0 .

In the opposite limiting case $\bar{\gamma} \gg 1$ we find from Eq. (7)

$$F(\bar{\gamma}) = \ln 2\bar{\gamma},$$

and the expression for the probability W is

$$W = (\mathcal{E}/(2\omega))^{2n} / 2^n. \quad (9)$$

Like Eq. (4), it depends only on the field intensity by a

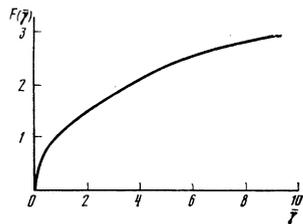


FIG. 2. The universal function $F(\bar{\gamma})$ of the adiabatic parameter $\bar{\gamma}$, which determines the probability of many-photon ionization in a stochastic electromagnetic field.

power law. The proportionality constant in the power law is changed.

Let us now turn to the calculation of the statistical factor G , defined as

$$G = W/w = \exp[2n(f(\gamma) - F(\bar{\gamma}))], \quad (10)$$

in which formula we set $\mathcal{E} = \bar{\mathcal{E}}$. Let us first examine the two limiting cases. Suppose $\gamma \gg 1$. Then, using Eqs. (9) and (4), we find from Eq. (10)

$$G = (n/e)^n. \quad (11)$$

According to Stirling's formula, to exponential accuracy this is equivalent to $n!$. This result is a well known one. The yield of a many-photon process caused by radiation with Gaussian statistics is larger by a factor $n!$ than the yield produced by one-mode (coherent) radiation¹⁶⁻¹⁸ with the same mean intensity. This result has been confirmed experimentally for two-photon¹⁹ and for five-photon¹⁵ processes.

Let us now consider the opposite limiting case $\gamma \ll 1$ (and *a fortiori* $\bar{\gamma} \ll 1$), for which the transition is of the tunnelling type. From Eq. (10) together with (3) and (8) we get

$$G = \exp[4(2E_0)^{3/2} / 3\mathcal{E}]. \quad (12)$$

In view of the condition $\mathcal{E} \ll \mathcal{E}_{at}$ we have $G \gg 1$ also in the tunnelling case. However, as can be seen by comparing Eqs. (11) and (12), the tunnelling statistical factor is much smaller than the many-photon statistical factor.

In the intermediate case the statistical factor is given by Eq. (10) in terms of the one-parameter functions $f(\gamma)$ and $F(\bar{\gamma})$ shown respectively in Figs. 1 and 2.

It must be emphasized that refinement of these formulas by taking the coefficient of the exponential into account is illusory, owing to the many inaccuracies present in attempts to get this coefficient in the original formula of Keldysh, Eq. (1). This means that attempts to calculate the general integral in Eq. (5) exactly, instead of by steepest descents, are excessively accurate and therefore superfluous.

In the experiment described in Ref. 15, for $n=11$ and $\gamma=5$ the value found was $\ln G = 12 \pm 2$. In this case $\bar{\gamma} = 1.5$, so that the asymptotic formula (11) for $n!$ must not be used, and we have to deal with the intermediate case, Eq. (10). Using Figs. 1 and 2, we find that the calculated value is $G = 12.5$, which is very close to the experimental value.

In another experiment,¹³ performed with an average radiation intensity of $0.98 \cdot 10^{12}$ W/cm², we find $\gamma = 9.3$ and $\bar{\gamma} = 2.8$. This situation falls in the many-photon region. By means of Figs. 1 and 2, or Eq. (11), which in this case is the same thing, we find $\ln G = 15.3$. This agrees with the experimental value,¹³ $\ln G = 15.87 \pm 0.69$. This agreement is to be regarded as a confirmation that the statistics of the laser radiation was Gaussian (the number of modes in the experiment was $N = 100$), as we

assumed in our calculation.

Owing to the nonexistence at present of experimental papers with $n > 20$, the following fact should be emphasized. Since

$$\gamma = \frac{1}{2n} \frac{\mathcal{E}_{at}}{\mathcal{E}}, \quad \bar{\gamma} = \frac{1}{2n^n} \frac{\mathcal{E}_{at}}{\mathcal{E}},$$

the conditions $\gamma, \bar{\gamma} \approx 1$ for which the ionization probability deviates from the power law are satisfied for

$$n \gg \mathcal{E}_{at}/2\mathcal{E},$$

in a monochromatic field, but for

$$n \gg (\mathcal{E}_{at}/2\mathcal{E})^{1/2},$$

in a random field; that is, for a lower value of n in the latter case. For the experiment of Ref. 15 we have $\mathcal{E}_{at}/\mathcal{E} = 110$; thus the inequality $\gamma \approx 1$ gives $n \approx 55$, whereas the inequality $\bar{\gamma} \approx 1$ gives $n \approx 15$. Accordingly, appreciable deviation of the ionization probability from the power law, characteristic of ordinary perturbation theory can be achieved more easily in a stochastic field than in a monochromatic field.

In conclusion we can state that the proposed theory contains simple universal one-parameter functions, by using which one can easily calculate probabilities of nonresonance many-photon ionization in a strong stochastic electromagnetic field, and also the corresponding statistical factors.

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¹⁾Kishinev State University.

- ¹L. V. Keldysh, Zh. Eksp. Teor. Fiz. 47, 1945 (1964) [Sov. Phys. JETP 20, 1307 (1965)].
- ²W. B. Delone, Usp. Fiz. Nauk 115, 361 (1975) [Sov. Phys. Uspekhi 18, 169 (1975)].
- ³J. S. Bakos, Adv. Electron. Electron Phys. 36, 57 (1974).
- ⁴S. A. Akhmanov and A. S. Chirkin, Statisticheskie yavleniya v nelineinoi optike (Statistical phenomena in nonlinear Optics), Izd. MGU (Moscow State Univ. Press), 1972.
- ⁵J. L. DeBethune, Nuovo Cim. 12B, 101 (1972).
- ⁶N. B. Delone, V. A. Kovarskiĭ, A. V. Masalov, and N. F. Perel'man, Preprint FIAN (Phys. Inst. Acad. Sci.) No. 120, 1978.
- ⁷S. S. Todirashku, Candidate's dissertation, Kishinev, 1979.
- ⁸V. A. Babenko, B. Ya. Zel'dovich, V. I. Malyshev, and A. A. Sychev, Zh. Eksp. Teor. Fiz. 61, 2270 (1971) [Sov. Phys. JETP 34, 1216 (1972)].
- ⁹R. V. Karapetyan, Izv. Vyssh. Uchebn. Zaved., Radiofiz. 28, 126 (1975).
- ¹⁰P. V. Elyutin, Pis'ma Zh. Tekh. Fiz. 2, 581 (1976) [Sov. Tech. Phys. Lett. 2, 227 (1976)].
- ¹¹S. S. Todirashku, Izv. Akad. Nauk Mold. SSR, ser. Fiz.-tekh. Mat. Nauk 1, 80 (1977).
- ¹²T. U. Arslanbekov, N. B. Delone, A. V. Masalov, S. S. Todirashku, and A. G. Faĭnshteĭn, Zh. Eksp. Teor. Fiz. 72, 907 (1977) [Sov. Phys. JETP 45, 473 (1977)].
- ¹³C. Lecompte *et al.*, Phys. Rev. A11, 1009 (1975).
- ¹⁴L. A. Lompre *et al.*, Phys. Rev. A15, 1604 (1977).
- ¹⁵T. U. Arslanbekov, Pis'ma Zh. Tekh. Fiz. 2, 536 (1976) [Sov. Tech. Phys. Lett. 2, 209 (1976)].
- ¹⁶V. A. Kovarskiĭ, Mnogokvantovye perekhody (Many-quantum transitions), Shtiintsa, Kishinev, 1974; Zh. Eksp. Teor. Fiz. 57, 1613 (1969) [Sov. Phys. JETP 30, 872 (1970)].
- ¹⁷B. R. Mollow, Phys. Rev. 175, 1555 (1968).
- ¹⁸C. Agarwal, Phys. Rev. A1, 1445 (1970).
- ¹⁹F. Shiga and S. Imamura, Phys. Lett. 25A, 706 (1967).

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Fine structure of the spectrum of depolarized light scattering in liquids

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It is shown that accounting for the correlation of simultaneous fluctuations leads to the appearance of terms with spatial velocity derivatives in the permittivity. As a consequence the permittivity is no longer a scalar and a fine structure appears in the spectrum of the depolarized light scattering.

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INTRODUCTION

The intensity of light scattered by a liquid consists of two essentially different parts (see, e.g., Ref. 1). The first is scattering by macroscopic fluctuations, i.e., the fluctuations of such hydrodynamic quantities as entropy or pressure. The second is scattering primarily by the anisotropy fluctuations and in general by the relative location fluctuations of the particles on microscopic scale. Both types of scattering give com-

parable contributions to the integrated scattering intensity, but are characterized by essentially different spectral distributions.

The microscopic fluctuations result in the appearance of non-shifted line (wing) the width of which $\Delta\omega_w$ is of the order of the characteristic frequency ν of microscopic movements in the liquid, $\Delta\omega_w \sim \nu = 1/\tau$ (for simplicity in the estimates below only simple liquids having no slowly relaxing parameters will be kept in mind).