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[12] The $^{12}C + ^{13}C$ system was thought to be the case most likely to exhibit phenomena similar to those inferred by Michaud and Vogt (Ref. 9) from their analysis of the then existing data (Refs. 2, 3, 5, and 7) for $^{12}C + ^{12}C$ and $^{12}C + ^{16}O$. However, none of the new experimental results presented in Fig. 1 seems to exhibit features similar to the $^{12}C + ^{12}C$ system which would suggest the presence of either broad shape resonances or absorption under the barrier (Ref. 9).

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High Resolution Threshold Photofragment Spectroscopy of $O_2^+(a\,^4\Pi_u \to f\,^4\Pi_g)$†

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A new technique of laser photofragment spectroscopy is introduced and applied to a study of transitions from the $v = 4$ level of the $a\,^4\Pi_u$ state of O_2^+ to predissociating levels of the $f\,^4\Pi_g$ state near the dissociation limit. A resolution of 0.5 MeV is obtained, and specific (Ω, J) levels of the a state are identified. Much higher resolution is possible.

Laser photofragment spectroscopy[1-5] has recently become an established technique for the study of the potential curves of molecular ions. With this technique, one crosses a fast beam of ions (1–10 keV) with polarized photons from a laser and measures the energy spectrum of the photofragment ions produced in photodissociation processes. By rotating the laser polarization, one obtains the angular distribution of these photofragments. There are well-established methods[1-6] with which one can use this information to determine the vibrational spacings of ground, metastable, and predissociative excited states, transition symmetries, molecular bond energies, and potential curves of dissociative states. This technique is limited, however, by the available resolution in the kinetic energy of the ejected photofragments. Even though the center-of-mass energies of the photofragments are greately magnified in the laboratory reference frame, the resulting resolution in the c.m. system for existing experiments[2-4] is about 50 meV, for separation energies W near 200 meV. However, when the dissociative process yields photofragments with W near zero, two processes can occur, both leading to a resolution substantially better than 1 meV, thus allowing study of the rotational and fine-structure states.

Consider the potential curves shown in Fig. 1. These curves are drawn to represent the $a\,^4\Pi_u$, $b\,^4\Sigma_g^-$, and the recently identified[3] $f\,^4\Pi_g$ states of O_2^+. Transition I represents a photon-induced transition between a specific level of the a state and the f state very near the dissociation limit. We assume here that the f state does not present a barrier to the dissociation. In typical experiments[2-5] which use magnetic energy analyzers for the photofragment energies, if one adjusts the magnetic field so that the "window" ΔH is centered at the value of H corresponding to an apparent energy W_A equal to zero, the resolution in energy ΔW can be shown from the basic formula of mass spectrometry, $(HR)^2 = 2MT_0/e$, to be[7]

$$\Delta W = (\Delta H/2H)^2 T_0, \qquad (1)$$

where T_0 is the kinetic energy of the beam in the lab frame and we have assumed photofragments of equal mass. For a beam energy of 2000 V and a resolution in magnetic field, $\Delta H/H$, of 10^{-3}, one obtains $\Delta W = 0.5$ meV. This expression applies only to photofragments ejected along the beam direction; and in general they can be ejected with a distribution between the square of the cosine and nearly isotropic with respect to the beam direction. This will decrease the resolution to about 0.8 meV full width at half-maximum for a cosine-squared distribution and 2 meV for an isotropic distribution. Due to the ΔH^2 term in Eq. (1), increasing the resolution of the magnet by a factor of 5 yields a resolution greater than 0.1 meV.

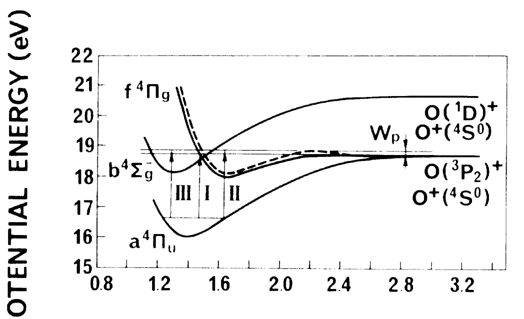

FIG. 1. Potential energy curves for three excited states of O_2^+.

Consider now an attempt to observe direct dissociation by transition I for $W_A = 0$. As the laser is tuned, one expects to observe photofragments each time the photon energy corresponds to a transition between a level of the lower state and the dissociation limit, with a resolution in energy of the initial level determined by the considerations just discussed. Thus one should observe individual rotational and fine-structure levels of the lower state. If the magnetic field is adjusted to observe W_A unequal but near to zero, any previously observed peaks should be shifted to larger photon energies, with substantially poorer resolution.

Consider now transition II in Fig. 1. It connects a level of the a state with a rotationally excited quasibound level of the f state. This level can dissociate after some time by tunneling through the centrifugal barrier. Rotational predissociation of ions has been studied in some detail[8] and the same considerations apply here. Even though a large number of rotational levels may be excited, in a given experiment one will observe only those which predissociate within a given lifetime window, 0 to 10^{-6} sec in the present experiment.

The photofragments for the rotationally predissociated level shown will appear with an energy corresponding to W_p in Fig. 1. Thus, if the energy-analyzer window includes W_p and the lifetime of this level is less than 10^{-6} sec, photofragments will be observed when the laser is on resonance between this level and some level of the a state. The resolution now does not depend on that of the energy analyzer but on the linewidth and stability of the laser, the lifetime of the dissociating level, and Doppler broadening caused by transverse energies in the beam. These effects can be made very small, and a resolution of 0.001 meV should be attainable. Obviously, if we change the energy analyzer to a different energy but such that the window still includes W_p, we will continue to see the photofragments at the same laser wavelength, but perhaps with a different intensity.

Exactly the same considerations apply if the f state has a true potential-energy barrier at large internuclear separation, except that now the direct dissociation discussed earlier cannot be observed at $W = 0$, but only for energies near or above the barrier height.

Consider now transition III between the a and b

states. The *b* state can now be predissociated by the *f* state. Again the resolution does not depend on the energy analyzer. If a predissociation of the *b* state occurs yielding photofragments with W near zero, one would expect to find also such a predissociation, with similarly well-resolved structure, for the next higher vibrational level, and perhaps for several levels beyond. Furthermore, for a given vibrational level one should observe photodissociation over a range of photofragment energies corresponding roughly to the rotational temperature of the ion.

We have previously[3] investigated the photodissociation of the $a\,^4\Pi_u$ state of O_2^+, and have shown that photofragment O^+ ions were produced under certain conditions with $W_A = 0$. Using the same experimental apparatus, we measured the production of O^+ photofragments as a function of photon energy in the energy range corresponding to dissociation from the $v = 4$ level of the *a* state with photofragment energy near zero. The magnetic energy analyzer was first set to $W_A = 0$. Under this condition, the collection efficiency is maximum for photofragments with energies between 0 and 0.5 meV, and decreases for higher energy in a manner depending on the (unknown) angular distribution of the photofragments. The experimental parameters corresponded to those used in the preceding illustrative discussion. A flashlamp-pumped dye laser with rhodamine 6 G was used.

The results are shown in Fig. 2. The photofragment count rate is corrected for the changing photon flux, as the laser is tuned. The energy scale gives the photon energy above or below that amount required to dissociate from the center of the four fine-structure levels of the *a* state with no rotational energy to the lowest dissociation limit, 2.141 eV. Thus peaks appearing at negative energy values in Fig. 2 correspond to higher rotational and fine-structure levels of the *a* state.

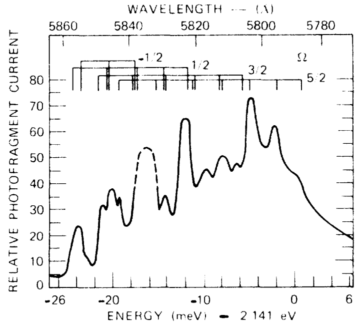

FIG. 2. Threshold photofragment spectrum for $O_2^+(a, v=4\rightarrow f)$. The dashed part of the spectrum contains at least three unresolved peaks. Above the spectrum (Ω, J) values calculated assuming $W_p = 4$ MeV are indicated. For each Ω, J begins with 9.5 at the right, and increases (in unit steps) to the left.

In order to choose between the three possible types of transitions discussed above, we made other experimental tests in the region between 5820 and 5860 Å. First, in order to determine the range of kinetic energies W to which these photofragments correspond, spectra in kinetic energy[3] W_A were taken at a number of wavelengths. We conclude that the W_p for these four peaks was 4 ± 2 meV. Second, in order to distinguish between direct dissociation and predissociation, the wavelength location of these peaks was investigated at various W_A between 0 and 8 meV. The positions remained fixed with approximately the same resolution, while the intensity decreased for $W_A > 2$ meV, consistent with $W_p = 4 \pm 2$ meV.

Finally, the effect of changing the direction of polarization of the laser was tested. In contrast to experiments at higher W,[2,3] essentially the same spectra were obtained with light polarization either parallel or perpendicular to the ion flight direction.

We can now draw several conclusions concerning the observed photodissociation. (i) Since the peaks remain at fixed wavelength as W_A is changed, we observe a predissociation, not a direct dissociation. (ii) Since the photofragments have very low kinetic energies (4±2 meV), the $f\,^4\Pi_g$ state does not have a large barrier against dissociation. A recent *ab initio* calculation[9] of this potential yielded a barrier of 47 meV. However, using this calculated potential, we found by numerical integration of the Schrödinger equation that one bound vibrational level exists above the dissociation limit, at 22 to 35 meV for rotational quantum numbers $R = 0$ to 12, the corresponding lifetimes varying from 4.4×10^{-6} to 5.5×10^{-8} sec. (iii) Since the photofragment kinetic energies are confined to a narrow range, it is unlikely that the $b\,^4\Sigma_g^-$ state is participating. Since the $v = 4$ vibrational level of the *b* state lies 10 meV below the dissociation limit, we would expect to see a maximum in the number of photofragments at about $W_A = 10$ meV since this would correspond to the peak of the rotational distribution at 20 meV, and substantial numbers of photofragments out to 25 meV. As an additional check, an extensive effort was made to observe predissociation of the $v = 5$ level of the *b* state. This predissociation, if it occurred, would be easy to distinguish from the direct dissociation.[3] No such predissociation was observed.

It should be pointed out that the "resolution" referred to for transitions II and III is for the separation of two peaks such as those in Fig. 2. Determination of the precise location of the upper dissociating level depends on the measurement of W_p. Although predictable apparatus improvements should allow determination of W_p to within ± 0.1 meV, we have here only made the relatively crude determination of 4 ± 2 meV. However, using $W_p = 4$ meV, the energies of the peaks corresponding to the various (Ω, J) levels of the $v=4$ level of the a state can be calculated[10] since the rotational and fine-structure constants for this state are known.[11] The results of such a calculation are compared with the observed peak location in Fig. 2. For each Ω listed, the appropriate J values begin at 9.5 at the short-wavelength end and increase to $J = 15.5$ for $\Omega = \frac{5}{2}$, 14.5 for $\frac{3}{2}$, 13.5 for $\frac{1}{2}$, and 11.5 for $-\frac{1}{2}$. A surprisingly close correspondence to the peak locations is observed, considering the ± 2 meV uncertainty in W_p. If one assumes photofragment energies of 2 or 6 meV, one can obtain a reasonable comparison with the observed peaks for J levels decreased or increased by 1, although it is substantially poorer than that shown in Fig. 2. Changing J by ± 2 is completely inconsistent with the observed W_p.

Transitions between the a and f states will obey the selection rules $\Delta J = 0, \pm 1$, and (not strictly) $\Delta\Omega = 0$. This fact, combined with the narrow range of observed W_p and the instrumental lifetime window, carries information about the barrier against dissociation. Unfortunately, to use this information effectively, one needs to know with better precision (± 0.1 meV) the W_p value for each peak in the spectrum. As suggested above, such precision is attainable. At present, neither the nature of this barrier, nor its location, nor the assignment of the participating upper quasibound states can be given. However, we may point out that (i) the height of this barrier is substantially less than 47 meV, but greater than zero, and probably near 5 meV; (ii) we apparently observe only processes leading to the lowest fine-structure state of the fragments, i.e., $O^+(^4S_0) + O(^3P_2)$; (iii) this choice of final state almost certainly influences the tunneling process; and (iv) if the barrier is located at large enough internuclear distances, the spin-orbit coupling operator will mix the $f\,^4\Pi_g$ state with other doublet, quartet, and sextet states connected with the same fragment state. All of these issues can be investigated in more quantitative detail after an improvement in the resolution of the apparatus.

We thus conclude that the (Ω, J) values indicated in Fig. 2 are the ones observed in the spectrum, although, because of the ± 2 meV uncertainty in W_p, the correspondence between (Ω, J) levels and observed peaks may not be exactly as indicated. Furthermore, the photodissociation occurs via the rotational predissociation of the $f\,^4\Pi_g$ state.

The resolution observed in the spectrum of Fig. 2 is at best 0.5 meV, substantially worse than the 0.07 meV indicated by the expected linewidth of 0.2 Å of the laser. However, this is the stated linewidth of the manufacturer; it was not verified. Furthermore, the stability in wavelength of the laser from flash to flash is unknown; and the mechanical tuning mechanism allows wavelength tuning reliably only in steps no smaller than 1 Å. These deficiencies in the operation of the laser can, of course, be easily remedied.

As demonstrated here, threshold photofragment spectroscopy has a resolution two orders of magnitude better than that obtained in normal photofragment spectroscopy. Predictable improvements suggest that for a direct dissociation a resolution of 0.1 meV should be easily attainable, and for a tunneling predissociation 0.001 meV. A number of molecular ions are known to have potential curves which should photodissociate at $W_A = 0$; for example, OH^+, CH^+, Ar_2^+, and F_2^-. This technique should thus be widely applicable to the spectroscopy of molecular ions.

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Crossover Dimensions for Fully Developed Turbulence

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The analytic continuation of homogeneous, isotropic turbulence in nonintegral dimensions d is not realizable for $d<2$ because the energy spectrum generally becomes negative. Recent arguments, in favor of an $\frac{8}{3}$ crossover dimension below which the Kolmogorov 1941 theory is exact, are found questionable. The existence of a crossover dimension d_c (≈ 2.03) at which the direction of the energy cascade reverses is supported by a second-order closure calculation.

The existence for critical phenomena of crossover dimensions d_c above which the mean-field theory is exact, and all the recent work on $d_c - d$ expansions lead one naturally to ask the same questions for fully developed, incompressible turbulence, a problem generally believed to be at least as difficult as critical phenomena.[1-3] For homogeneous isotropic turbulence with Gaussian initial conditions and forcing, the energy spectrum $E(k)$ is, in principle, a well-defined functional of the forcing and initial spectra, which may be continued analytically in noninteger dimensions using, for example, the formal Reynolds number expansion.

(I) In a recent Letter, Forster, Nelson, and Stephen[4] analyze an infrared problem for $d \approx 2$ and find, for $d<2$, a nontrivial fixed point. We do believe that $d=2$ is a crossover dimension, but only in the sense that, for $d<2$, the homogeneous isotropic turbulence problem is no longer meaningful. Indeed, we have shown that for $d<2$ the energy spectrum, if initially positive, will usually become negative for arbitrarily small times. The essence of the proof is that (i) for small times the quasi-normal approximation is exact to $O(t^3)$ and (ii) the quasi-normal expression of the transfer has an emission coefficient, usually denoted[5-7] a_{kpq} which when $p=q$ becomes negative for $d<2$, whereas it is always positive for $d \geqslant 2$. The realizability [in the sense $E(k) \geqslant 0$] of the analytic continuation into nonintegral dimension $d>2$ remains open in general, but can be settled within the framework of second-order closures.

(II) An $\frac{8}{3}$ crossover dimension for intermittency corrections to the Kolmogorov 1941 theory (K41) has been proposed independently by Nelkin[2] and de Gennes.[8] The de Gennes derivation is parallel to the classical Ginsburg argument for critical phenomena: Denoting $\epsilon(x)$ the local dissipation $\nu(\nabla v)^2$ and assuming K41, we have for separations $\vec{s}$ lying in the inertial range, by dimensional analysis ($\bar{\epsilon}$ is mean energy dissipation),

$$\langle[\epsilon(\vec{s})-\bar{\epsilon}][\epsilon(\vec{0})-\bar{\epsilon}]\rangle \sim \nu^2 \bar{\epsilon}^{4/3} s^{-8/3}.$$

Since the volume element goes like $s^{d-1} ds$, the mean square dissipation fluctuation has an infrared divergence if $d > \frac{8}{3}$.

Our point is that $\nu(\nabla v)^2$ is a dissipation-range quantity and that its inertial-range fluctuations are irrelevant. If, instead, we use the nonlinear expression of the rate of transfer of energy $\epsilon(x) \sim \vec{v} \cdot \nabla(v^2/2 + p)$, we find

$$\langle[\epsilon(\vec{s})-\bar{\epsilon}][\epsilon(\vec{0})-\bar{\epsilon}]\rangle \sim \bar{\epsilon}^2 s^0,$$

and the crossover dimension becomes zero. Alternatively, we can argue that the inertial-range properties should not be changed if the dissipative term in the Navier-Stokes equation, namely $\nu\nabla^2$, is replaced by $-\nu(-\nabla^2)^\alpha$, where α is called dissipativity. Indeed it is shown[9] for a certain model equation that the inertial-range behavior is unaffected by the dissipativity in the limit of zero viscosity, as long as $\alpha > \alpha_{\mathrm{cr}} = (n-1)/2$, where n is the exponent of the inertial range. It is clear that in de Gennes's (and also Nelkin's) derivation, the crossover dimension is dissipativity-dependent (e.g., for $\alpha = 2$ we find $d_c = \frac{20}{3}$ but for $\alpha = \alpha_{\mathrm{cr}} = \frac{1}{3}$, we recover $d_c = 0$ again). All this suggests that K41 is invalid in any realizable ($d \geqslant 2$) dimensions.