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⁹C. N. Yang, Phys. Rev. Lett. <u>38</u>, 1377 (1977); we follow the notations and conventions of this paper, " $\stackrel{\bullet}{=}$ " is used for equations valid for real values of x_1, x_2, x_3 , and x_4 .

¹⁰Note that by simple counting J' is overdetermined by Eqs. (14a) and (14b). This often happens in the BT, e.g., the BT for sine-Gordon equations, and nonlinear σ model [K. Pohlmeyer, Commun. Math. Phys. <u>46</u>, 207 (1976)].

¹¹Equation (15) follows from Eqs. (14a) and (14c) provided α is a real constant and $J'J^{-1}=JJ'^{-1}+B$ where B is a constant matrix. The consistency of the equation $J'J^{-1}=JJ'^{-1}+B$ requires $[B,JJ'^{-1}]=[B,J'J^{-1}]=0$. Since $J'J^{-1}$ in general depends on y, \overline{y} , z, and \overline{z} , we must take $B=\beta I$ where I is the identity matrix and β is an arbitrary constant. Taking the trace of Eq. (14b) and using Eq. (14c) show that β must be real.

¹²P. J. McCarthy, Lett. Math. Phys. <u>2</u>, 167, 492 (1978); the classical Bäcklund transformation for the sine-Gordon equation is, in fact, "strong."

¹³To compare our results with those of Pohlmeyer, one needs the following definitions and identities:

 $q_a \equiv g_{ab} q^b$

$$g_{ab} = \operatorname{diag}(+1, -1, -1, -1) \quad (a, b = 0, 1, 2, 3),$$

$$q^{2} \equiv q^{a}q_{a} \equiv (q^{0})^{2} - \vec{q} \cdot \vec{q};$$

$$J \equiv q^{a}\alpha_{a} \equiv q^{0}I + \vec{q} \cdot \vec{\sigma}; \quad J \equiv q^{2}J^{-1} = q^{0}I - \vec{q} \cdot \vec{\sigma};$$

$$J' \equiv q'^{a}\alpha_{a}; \quad J'' \equiv q''^{a}\alpha_{a};$$

$$q \cdot q' \equiv q^{a}q_{a}'; \quad [q, q', q'']^{a} \equiv \epsilon^{abcd}q_{b}q_{c}'q_{a}'';$$

$$JJ' + J'J = JJ' + J'J = 2q \cdot q'I;$$

$$JJ' + J'' = \{(q \cdot q')q''^{a} + (q' \cdot q'')q^{a} - (q \cdot q'')q'^{a}$$

 $+i[q,q',q'']^a$ α_a .

¹⁴By way of an example, we use transformation $[\alpha\beta]$ twice (i.e., $J \rightarrow J' \rightarrow J''$) on the trivial (i.e., vacuum $F_{\mu\nu} \equiv 0$) SU(2) gauge field solution J = I. In the first step, we choose $\beta \equiv 0$ and take $J' = \sigma_1$ which is a trivial solution of Eq. (11) for SU(1, 1) gauge group. In the next step, we let $\alpha \equiv 0$ and $\beta \equiv 2\gamma$ and parametrize J'' [belonging to SU(2) gauge group again] as

$$J'' = \begin{pmatrix} \frac{1}{\varphi} & \frac{\overline{\rho}}{\varphi} \\ \frac{\rho}{\varphi} & \varphi + \frac{\rho\overline{\rho}}{\varphi} \end{pmatrix}, \quad \overline{\rho} \stackrel{\bullet}{=} \rho^{\dagger}, \varphi \stackrel{\bullet}{=} \text{real.}$$

Equation (14b) then implies $\rho + \overline{\rho} = 2\gamma \varphi$. The differential equation (14a) $J'^{-1}J_{y'} - J''^{-1}J_{y''} = (J'^{-1}J'')_{\overline{z}}$ for $J' = \sigma_1$ implies $\varphi_y = -\rho_{\overline{z}}$, $\varphi_z = \overline{\rho_{\overline{y}}}$, $\varphi_{\overline{y}} = -\rho_z$, $\varphi_{\overline{z}} = \rho_y$ which, with the constraint equation $\rho + \overline{\rho} = 2\gamma\varphi$, have the solution $\varphi = (2\delta)^{-1}(f+\overline{f})$, $\rho = f + \epsilon\varphi$, $f \equiv f(\epsilon z + \overline{y}; \epsilon y - \overline{z})$, $\overline{f} \equiv f^{\dagger}$, $\delta = (1 + \gamma^2)^{1/2}$, $\epsilon = \gamma + \delta$, where *f* is an arbitrary function. The action density corresponding to this solution is $S = -\frac{1}{2} \Box \Box \ln \varphi = -\frac{1}{2} \Box \Box \ln (f+\overline{f})$ which for any nontrivial (i.e., $S \neq 0$) *f* always diverges on a three-dimensional hypersurface of four-dimensional Euclidean space.

Deflection of a Na Beam by Resonant Standing-Wave Radiation

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A symmetric deflection of a Na beam by resonant standing-wave radiation has been observed. The combination of absorption and stimulated emission of the two oppositely traveling waves ω_+ and ω_- results in a net deflection of the beam. The observed magnitude of the beam deflection and its dependence on the radiation power agree with a calculation based on the random-walk picture that the two traveling waves ω_+ and ω_- are utilized randomly in the stimulated-emission and absorption processes.

The deflection of an atomic beam by resonant traveling-wave radiation ω_+ has been well demonstrated experimentally.^{1,2} In these experiments, unfocused resonant radiation from an atomic emission source¹ or a laser² was impinged from an orthogonal direction on an atomic beam. An atom, on repetitively undergoing an absorption and a spontaneous emission, experiences a deflection in the direction of ω_+ . The induced emission process does not increase the deflection because the photon momentum transferred to the atom is opposite to the direction of ω_+ .³

It has been proposed^{4, 5} that if resonant radiation traveling in both directions, ω_+ and ω_- , is used, the stimulated emission process will reinforce the effect of the absorption and deflect the beam further. It has also been pointed out⁶ that a Raman process in a standing wave,⁷ which was later named a velocity-tuned three-photon process,⁸ has a momentum transfer of three units in a single step. Such a process has been observed in a CO_2 -laser cavity^{8, 9} and it has been noted that a velocity-tuned (2l+1)-photon process has a momentum transfer of 2l + 1 photons in one direction. However, this would not apply to very large *l*. In this paper we study experimentally this extreme case by using an intense resonance standing wave and a Na beam. This experiment is an optical analog of the rf transverse Stern-Gerlach experiment¹⁰ but the result is qualitatively different as described below. This is also an atomic analog of the Kapitza-Dirac effect¹¹ in which the deflection of an electron beam due to standing-wave radiation is considered.¹² However, the use of resonant radiation force rather than the Compton scattering makes the present effect much easier to observe. An ingenious experiment of atomic-beam deflection by copropagating resonant radiation has also been reported.¹³

The theory for the transfer of momentum to atoms by a resonant standing wave has been actively studied recently by Kazantsev,¹⁴ Stenholm,¹⁵ and Cook and Bernhardt.^{16, 17} The basic idea of our experiment can be seen most clearly by applying the photon momentum picture to the two traveling waves ω_+ and ω_- interacting with an atomic beam (Fig. 1). Four induced processes are possible: absorption of ω_+ and ω_- which we denote ω_{+}^{\dagger} and ω_{-}^{\dagger} , respectively; and induced emission of ω_+ and ω_- which we denote ω_+^{\dagger} and ω_{+}^{\dagger} , respectively. The processes ω_{+}^{\dagger} and ω_{-}^{\dagger} push an atom in the +x direction, while ω_+^{\dagger} and ω_{-}^{\dagger} push it in the -x direction. An atom makes transitions between the two levels alternately absorbing and emitting a photon $\hbar \omega$ at a rate given by the Rabi "flopping frequency" of $\mu E/h$. Using a moderate laser power of 100 mW focused to 0.1mm radius we calculate the Rabi frequency to be ~1 GHz for the $2^2 P_{3/2} - 2^2 S_{1/2}$ transition of Na, which is much larger than the spontaneous emission rate $\frac{1}{2}\tau$ with $\tau = 1.63 \times 10^{-8}$ sec.¹⁸ Using an average Na atomic velocity of v = 900 m/sec in the beam and the size of the interaction region of l = 0.2 mm, we find that an atom undergoes induced processes $n = \mu E l/hv \sim 200$ times while the spontaneous emission occurs only several times during the transit time $t \sim 2 \times 10^{-7}$ sec.

The net transfer of photon momenta to an atom depends on which combinations of ω_+ and ω_- are used in the stimulated processes: (a) The maxi-

mum deflection in the +x direction would occur if ω_{+}^{\dagger} and ω_{-}^{\dagger} occur alternately, and that in the -x direction if ω_{+}^{\dagger} and ω_{-}^{\dagger} occur. The total deflection of the beam then would be proportional to *n* and thus to the Rabi frequency. (b) The minimum deflection would occur if an atom keeps interacting with only one standing wave. (c) If in the alternating absorption and stimulated-emission processes it is random whether ω_{+} or ω_{-} is involved, then the net deflection results from a random walk and has a Gaussian shape with a half-width which is proportional to \sqrt{n} .

The average deflection δ of the beam for the three cases can be estimated as

$$\delta \sim \frac{\hbar\omega}{c} \frac{1}{mv} Lf(n) \tag{1}$$

with f(n) = n for case (a), $f(n) = \frac{1}{2}$ for case (b),³ and $f(n) = \sqrt{n}$ for case (c), where L is the length of the free-flight region and m is the mass of the atom. For $n = \mu E l/hv \sim 200$ as estimated earlier, there is an order-of-magnitude difference between the three cases.

The essential elements of our apparatus can be seen in Fig. 1. The Na beam was produced by heating Na metal to 380°C in an iron oven with a hole of 0.1 mm. The collimator aperture was 30 cm from the oven and the opening was adjusted by micrometers attached to the four blades of the collimator such that the beam size at the ionizing wire was 0.25 mm vertically; horizontally the beam was two to three times wider. The slit in front of the ionization wire detector was 0.1 mm wide in the vertical direction and its vertical position was swept by a motor for scanning the beam profile. The detector was a hot-wire ionizer of 0.125 mm tungsten grown from W(CO)₆. The Na ions were collected on an electron multiplier after a crude mass spectrometer separated them from the background potassium ions from the wire. The Na beam was chopped at 30 Hz with toothed wheel driven by a synchronous motor. The output from the electron multiplier was processed with a lock-in amplifier and displayed







FIG. 2. Na-beam intensity profiles. (a) Undeflected beam (full width at half maximum ~ 0.25 mm). (b) Symmetric deflection of the Na beam due to resonant standing-wave radiation. Rabi frequency was ~ 2.1 GHz. (c) Laser-chopped signal which gives a difference between (a) and (b).

on a recorder. A stabilized Spectra Physics 580A dye laser provided single-mode resonant radiation with a power of up to 100 mW. The laser radiation was focused on the atomic beam with a lens of 10-cm focal length at a position 5 cm from the collimator slit and reflected by a concave mirror. The polarization of the laser electric field was horizontal and perpendicular to the atomic beam (along the y axis in Fig. 1). The free-flight distance of Na atoms after the interaction region was 75 cm.

A typical result of the deflection experiment is given in Fig. 2. In these experiments the laser frequency was fixed at the maximum of the sodi- $\operatorname{um} D_2$ line and the slit in front of the detector wire was swept. Because of the large Rabi frequency of ~1 GHz, the setting of the laser frequency was not critical. Figure 2(a) shows a trace of the Na beam without laser radiation; the half width at half maximum is about 0.13 mm. Figure 2(b) shows the symmetric deflection of the beam due to the standing wave. The large reduction of the peak and the broadening in the tail of the beam is clearly seen. The laser power was 100 mW and the frequency was set at the D_2 line. Although no attempt was made to measure the size of the interaction region directly, some idea was obtained from the magnitude of the Rabi frequency. The latter was found, by sweeping the laser frequency, to be ~ 2.1 GHz which indicated that the focusing of the dye laser was to



FIG. 3. Dependence of the deflection 2δ (full width at half maximum) on the radiation power. Black circles represent "raw" values without correction. White circles are corrected values. Curve *a* represents calculated values based on the model (a) in the text and curve *c* for the model (c). The model (b) gives a straight line very close to the abscissa.

better than 0.1 mm. When the reflecting mirror is blocked so that the atomic beam interacts with a focused traveling wave, a trace similar to Fig. 2(a) is obtained with a small shift in the direction of the traveling-wave radiation. Since such a shift was too small to be seen in an illustration, this case is omitted in Fig. 2.

The results in Figs. 2(a) and 2(b) are the "raw" data in which contributions from that portion of the Na beam which was not affected by the laser beam are also included. In order to exclude the contribution from this portion of the Na beam, the laser radiation rather than the atomic beam was chopped (at 30 Hz) and the signal was processed in a lock-in amplifier. This is equivalent to taking a difference between the traces (a) and (b) in Fig. 2. The result is shown in Fig. 2(c). The deflection (2δ) of the beam was measured from Fig. 2(c) by fitting a Gaussian curve to the lobes of the trace, as shown by the dotted line.

The deflection of the Na beam has been measured as a function of dye-laser power from 5 to 100 mW. The results are plotted in Fig. 3. The black circles represent values which are directly obtained from the measurements. The scatter is mainly due to unavoidable slight misalignment of the laser radiation with respect to the atomic beam for different runs. If we normalize the set of measurements in a given run by adjusting one value of the set to the theoretical curve, we obtain values represented by the white circles in Fig. 3. The theoretical curves for the cases (a) and (c) described above are also shown in Fig. 3. In drawing the latter, the laser electric field was estimated from the power broadening. It is seen that both the absolute value of the deflection and the power dependence of the deflection indicate that case (c) is occurring.

One complication arises due to the fact that the Na atom has two hyperfine levels with F = 2 and 1 in the ground state, and the optical transitions starting from these levels are separated by $\nu_1 - \nu_2 \sim 1.77$ GHz.¹⁹ Therefore if we set the laser frequency to the transition from the F = 2 level, the other transition is off resonant. The deflection of the Na atom in the F = 1 level is then estimated from $n = (\mu_1 E/h)^2 [(\nu_1 - \nu_2)^2 + (\mu_1 E/h)^2]^{-1/2}t$ rather than from $n = (\mu_2 E/h)t$.

In conclusion, our observation agrees with the random-walk picture, i.e., case (c). This is in contrast to the stimulated processes in traveling-wave radiation where case (b) applies and to the velocity-tuned multiphoton processes where, since an atom is "forced" to interact with ω_+ and ω_- alternatively, case (a) applies.

Contrary to our intuitive photon-momentum picture, most of the previous theoretical papers¹⁴⁻¹⁷ use a classical treatment for the interaction between the standing-wave radiation field and the atomic beam. Cook and Bernhardt¹⁶ have obtained a result which is formally identical to the Fraunhofer diffraction of a plane wave by a sinusoidal phase grating, i.e., the atomic beam is diffracted by the periodic amplitude of the standing wave (the picture originally proposed by Kapitza and Dirac¹¹). It is interesting to note that their theory predicts an average momentum transfer which agrees with our case (a) (apart from a constant factor). Essentially the same result is obtained from the classical picutre of Kazantsev^{14, 20} by using $\Delta p_x = Ft = -t \partial U(x) / \partial t$ with a standing-wave potential energy of $U(x) = -\mu E \cos kx$. Such calculations agree with the result of the transverse Stern-Gerlach experiment¹⁰ but differ both in the order of magnitude and the field dependence from what we observed.

Cook and Bernhardt considered a case where the effect of recoil and Doppler frequency shifts are not negligible and obtained a maximum momentum transfer [Eq. (27) of Ref. 16]; their result has the same field dependence as our case (c) but the magnitude of the deflection is larger than our observation by a factor of ~3. Kazantsev *et al.*²⁰ gave a similar formula considering the randomness of each atomic position with respect to the standing wave. For an ideal situation of a completely collimated beam $(\Delta p_x = 0)$, such a consideration is irrelevant, but for our atomic beam of $\Delta p_x / p \sim 3 \times 10^{-4}$, the position uncertainty $\Delta x \sim \hbar / \Delta p_x \sim 100$ Å is still smaller than the wavelength of the radiation.

It should be noted that in our random case (c) which agrees with the observation, the effect of coherence is completely neglected. Whether a more complete theory will actually predict effectively such behavior or whether our result has been brought about by some randomizing process (such as spontaneous emission, weak collisional interaction, or a nonideal experimental setup) remains to be seen. Finally, we note that in order to test the features predicted by Cook¹⁷ or even the finer diffraction "pattern" of the beam, an atomic beam with much higher collimation is needed.

We have profited from discussions with A. Bambini, A. H. Bernhardt, M. Bloom, R. J. Cook, and U. Fano about the interpretation of our experimental results.

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Effects of Shape Resonances on Vibrational Intensity Distributions in Molecular Photoionization

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We report striking non-Franck-Condon vibrational intensity distributions associated with the shape resonance in the 5σ photoionization channel of CO. This example confirms the recent theoretical prediction that shape resonances will couple significantly with vibrational motion, leading to different resonance energies and profiles, and non-Franck-Condon intensities in alternative vibrational channels. Analogous effects are expected in connection with the widespread occurrence of shape resonances in both innershell and outershell molecular photoionization spectra.

The prominent role of shape resonances in molecular photoionization has gained wide recognition in the last few years. Their identification¹ in the innershell and outershell spectra of a growing, diverse collection of molecules has led to the study of their role in partial photoionization cross sections^{1d, 1e, 2} and photoelectron angular distributions.^{1d, 3} Recently, a new manifestation of shape resonances has been predicted⁴ by theoretical studies of the effects of vibrational motion on the quasibound states. In particular, shape resonances were found to induce significant coupling between the escaping photoelectron and the vibrational motion of the nuclei which is manifested as large, energy-dependent deviations from Franck-Condon (FC) vibrational intensity distributions over a broad spectral range encompassing the resonance. In this Letter, we present the first experimental evidence for this behavior in connection with the σ -type shape resonance in the

 5σ photoionization channel of CO. This class of phenomena will play a central role in vibrationally resolved photoelectron studies conducted over the broad and continuous wavelength range afforded by synchrotron radiation sources.

The effect arises⁴ from the quasibound nature of the shape resonance, which is localized in a spatial region of molecular dimensions by a centrifugal barrier. This barrier and, hence, the energy and lifetime (width) of the resonance are sensitive functions of internuclear separation and vary significantly over a range of R corresponding to the ground-state vibrational motion. In an adiabatic treatment, the net dipole amplitude for a particular vibrational channel is obtained by averaging the R-dependent dipole amplitude, weighted by the product of the initial- and finalstate vibrational wave functions at each R. Accordingly, transitions to alternative vibrational levels of the ion preferentially weight different