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Alternative Decay Channels of a CO^- Feshbach Resonance*

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By means of on- and off-resonance energy-loss measurements, we have observed the decay of the 10.04-eV resonance in CO to the $A\,^1\Pi$, $a\,^3\Pi$, and $a'\,^3\Sigma^+$ states. The measurements identify the resonance as a $^2\Sigma^+$ state of CO^-.

A resonance in scattering of electrons on CO has been discovered by Sanche and Schulz[1a] in transmission at ≈ 10.04 eV. Subsequent work has indicated that the resonance is observable and approximately isotropic in both elastic scattering[1b-3] and in inelastic excitation of the $v'=1$ vibrational level of the ground electronic state.[3] The purpose of this Letter is to report experimental observations of decay of this resonance into a number of vibrational levels of at least three different electronically excited states. These observations were made by two different experimental techniques, i.e., (a) by obtaining excitation functions for individual vibrational levels and (b) by observing the energy-loss spectra both on and off resonance. The information obtained in these measurements provides (a) a definitive classification of the resonance and (b) direct experimental evidence of configuration interaction occurring between electronic states which comprise the $a'\,^3\Sigma^+$ decay channel and the $b\,^3\Sigma^+$ parent Rydberg state.

The instrument used in these measurements was a hemispherical electron monochromator analyzer using copper hemispheres of 5.08 cm (2 in.) mean radius as the energy dispersing elements. The target gas cell was a molybdenum cylinder 1.27 cm (0.5 in.) in diameter, with an entrance aperture of 0.165 cm diam (0.065 in.) and 0.063-cm-diam (0.025-in.) exit apertures at $\theta = 0°$, 30°, 45°, and 90°. The gas pressure inside the cell was about 0.02 Torr, and about 1000 times less than that in the surrounding vacuum chamber. The entire monochromator section can be rotated about the scattering center in fixed angular steps of 5° from 0° to 100°. The scattered electrons are energy analyzed in the analyzer hemisphere and counted by an eighteen stage Be-Cu multiplier. Output pulses produced by the individual electrons are transmitted down a 50-Ω line to a fast counting chain and can be counted on a scaler and stored in an on-line computer or displayed on a count-rate meter connected to an X-Y recorder.

Figure 1 shows excitation functions corresponding to the $v'=2$, 3, and 4 levels of the $A\,^1\Pi$ state of CO along with a background trace taken at 8.45-eV excitation energy. These excitation functions as well as all other data reported here were obtained at a scattering angle of 45° with an instrumental resolution of less than 50 meV full width at half-maximum. The data in Fig. 1 do not represent "pure" excitation of the indicated

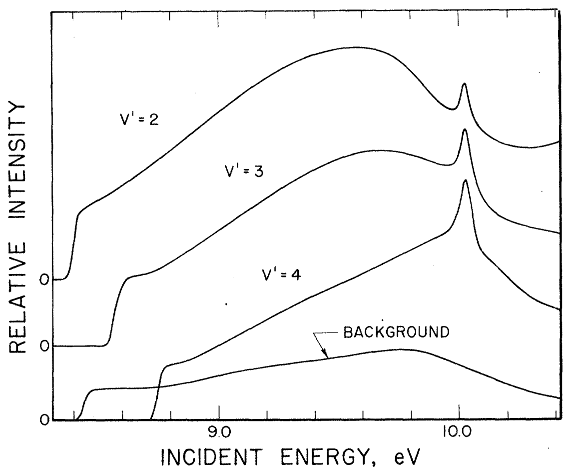

FIG. 1. Excitation functions for the $v'=2$, 3, and 4 vibrational levels of the $A\,^1\Pi$ state at 8.39, 8.57, and 8.73 eV, respectively. The background curve corresponds to an excitation energy of 8.45 eV. Noise has been suppressed in the curves shown, but is everywhere less than 10% of the height of the 10.04-eV peak above the nonresonant background.

states for two reasons. First, the instrumental resolution is insufficient to separate completely the indicated levels from neighboring levels of other electronic states. Second, there is a large background current as shown in the figure. Nevertheless, the resonance appears as a small resolved peak at 10.04 ± 0.02 eV as measured from the excitation threshold. Similar data have been obtained for a number of vibrational levels of the $A\,^1\Pi$, $a\,^3\Pi$, and $a'\,^3\Sigma^+$ states and will be reported in detail elsewhere. None of our results to date show any evidence of higher lying vibrational members of the 10.04-eV resonance. Similar measurements made of the excitation function for the $v'=1$ vibrational level of the ground state failed to reveal evidence of resonant structure even though this has been observed by others.[3] We attribute our failure to observe the resonance to a strong background of electrons due to elastic scattering in our $v=1$ ground-state excitation measurements.

In order to study the effect of the resonances on a number of excited levels simultaneously, energy-loss spectra were obtained for two values of incident energy, one corresponding to the resonance energy and another corresponding to 0.2 eV above it. The exact value of the resonance energy was defined by measuring the energy of the dip in elastic scattering. The results are shown in Fig. 2. Vibrational levels of the $a\,^3\Pi$, $a'\,^3\Sigma^+$, $d\,^3\Delta_i$, and $A\,^1\Pi$ states are marked by vertical lines at the top of the figure.[4] The energy scale of the figure is estimated to be accurate to about 5 meV.

The $a\,^3\Pi$ vibrational levels $v'=0$ to 6 can be clearly resolved. The intensity of each of these levels is enhanced about 20% in the "on-resonance" loss spectrum. The $a'\,^3\Sigma^+$ vibrational spectrum first becomes visible at 7.30 eV, where the $v'=3$ level appears as a shoulder to the right of the $a\,^3\Pi$, $v'=6$ level. The $a\,^3\Pi$, $v'=7$, 8 levels may be left and right shoulders, respectively, on the $a'\,^3\Sigma^+$, $v'=4$ and 5 peaks, but the $a\,^3\Pi$, $v'=9$

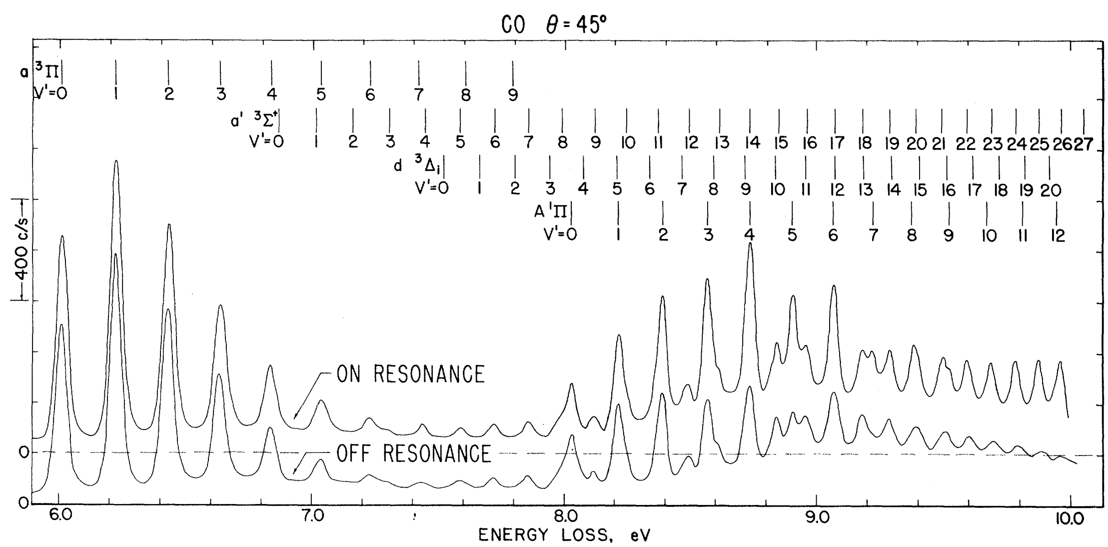

FIG. 2. Energy-loss spectra in CO at 45° for incident energies of 10.04 eV (on resonance) and 10.24 eV (off resonance). Energy levels for $a\,^3\Pi$, $a'\,^3\Sigma^+$, $d\,^3\Delta_i$, and $A\,^1\Pi$ states are shown at the top of the figure (Ref. 4).

TABLE I. Increase in the on-resonance loss peak intensity compared to the off-resonance intensity assuming only the $a'\,^3\Sigma^+$ and $A\,^1\Pi$ states are enhanced above 8.0-eV energy loss. These numbers have been corrected for ~10–20% variations in nonresonant scattering intensity between the resonance and 0.2 eV above the resonance. The overall total for $a\,^3\Pi$, $A\,^1\Pi$, and $a'\,^3\Sigma^+$ is 3020 counts/sec.

v'	$a\,^3\Pi$ (counts/sec)	$A\,^1\Pi$ (counts/sec)	v'	$a'\,^3\Sigma^+$ (counts/sec)	v'	$a'\,^3\Sigma^+$ (counts/sec)
0	75	15	3	...	15	20
1	95	60	4	10	16	40
2	75	105	5	10	17	40
3	60	150	6	10	18	40
4	45	270	7	15	19	60
5	35	240	8	15	20	70
6	25	210	9	15	21	80
7	10	130	10	15	22	100
8		100	11	20	23	110
9		70	12	20	24	130
10		40	13	20	25	160
11		10	14	20	26	180
Total	420 (14%)	1400 (46%)				1200 (40%)

level is not apparent. The succeeding $a'\,^3\Sigma^+$ levels can be followed either as separate peaks or as part of composite larger peaks all the way to the $v'=26$ level at 9.970 eV. The $a'\,^3\Sigma^+$ vibrational levels are weakly enhanced on resonance compared to off resonance until about $v'=16$, where they gradually appear more and more strongly. Their relative intensities may be obscured by the $d\,^3\Delta_i$ vibrational levels $v'=7$ to $v'=16$, which could contribute significant intensities to the peaks from 8.47 to 9.52 eV, and which fall within 30 meV of an $a'\,^3\Sigma^+$ level in that energy range. Above 9.52 eV the $d\,^3\Delta_i$ levels lie in the valleys between the loss peaks and there is no evidence of these levels on resonance.

The $A\,^1\Pi$ vibrational levels are the levels most enhanced by the resonance (see Table I). The vibrational levels $v'=0$–9 are clearly visible in the on-resonance spectrum, while the $v'=7$ and higher levels diminish rapidly in the off-resonance spectrum.

The $D\,^1\Delta$ state would not be expected to appear in the off-resonance spectrum until about $v'=6$ or higher, judging from the position of its potential curve with respect to that of the ground state.[4] There is no sign in either of the loss spectra of the $v'=5$, 7, 8, 9, 11, 12, 14, and higher levels at energies where they might appear unobscured by other peaks.

If we arbitrarily assign the increase in intensity occurring in the on-resonance spectrum above 8.0 eV to the $a'\,^3\Sigma^+$ and $A\,^1\Pi$ levels only, we would obtain the results shown in Table I for the distribution of the enhanced intensity. The total increase in the on-resonance inelastic intensity at $\theta=45°$ is roughly equal to the decrease in the elastic scattering intensity at resonance at $\theta=45°$.

The ground state of CO has the valence electronic configuration $1\pi^4 5\sigma^2$ ($X\,^1\Sigma^+$). Valence states are formed either by excitation of a 5σ or 1π orbital to form the configurations $1\pi^4 5\sigma 2\pi^1$ ($a\,^3\Pi$, $A\,^1\Pi$) or $1\pi^3 5\sigma^2 2\pi$ ($a'\,^3\Sigma^+$, $d\,^3\Delta_i$, $e\,^3\Sigma^-$, $I\,^1\Sigma^-$, $D\,^1\Delta$). The lowest Rydberg states, $b\,^3\Sigma^+$ and $B\,^1\Sigma^+$ ($1\pi^4 5\sigma 6\sigma$) (10.39 and 10.77 eV, respectively, above the $X\,^1\Sigma^+$ ground state), are formed by adding a 6σ (or $3s$) electron to the $1\pi^4 5\sigma^2\Sigma^+$ ground state of CO^+. The 10.04-eV resonance is interpreted[5] as the $v'=0$ component of a Feshbach-type resonance formed by binding an extra electron in a 6σ orbital to form the configuration $1\pi^4 5\sigma 6\sigma^2\,^2\Sigma^+$ of CO^-. This interpretation is similar to that recently proposed[6] for the analogous resonance in N_2^- ($^2\Sigma_g^+$ at 11.48 eV above the N_2 ground state and bound by ≈0.4 eV to the $E\,^3\Sigma_g^+$ of N_2). The similarity in shape of the resonance in elastic scattering at 45° (Fig. 1) with earlier measurements[3] at 30° and 90° supports this assignment. It is further supported by the fact that resonant enhancement is strong in the $a\,^3\Pi$ and $A\,^1\Pi$ states. If the electron orbital assignments given above were exact the only possible decay of a $5\sigma 6\sigma^2\,^2\Sigma^+$ resonance would be by a two-electron (Auger) process,[7] either to the $5\sigma^2\,^1\Sigma^+$ ground state or to the $5\sigma 2\pi$ $^3\Pi$ or $^1\Pi$ states.

Resonant effects are strongest for $v' = 0$ in the $X\,^1\Sigma^+$ ground state (elastic scattering), $v' = 1$ in $a^3\Pi$, and $v' = 4$ in $A^1\Pi$. This decay pattern would be expected from a $^2\Sigma^+$ resonance having approximately the internuclear separation of the $b\,^3\Sigma^+$ parent state of the resonance ($r_e = 1.12$ Å), which is approximately the same as that of the ground state ($r_e = 1.128$ Å) but smaller than that of the $a^3\Pi$ ($r_e = 1.206$ Å) and $A^1\Pi$ ($r_e = 1.235$ Å) states.

Decay into states of the $1\pi^3 5\sigma^2 2\pi$ configuration can occur only through configuration mixing of states of this configuration with states to which decay of a $5\sigma 6\sigma^2$ $^2\Sigma^+$ CO^- resonance is possible by a two-electron process or alternatively through configuration mixing of the resonant state itself with higher states of the same symmetry. In both cases the $^3\Sigma^+$ state is the most likely decay channel of the $1\pi^3 5\sigma^2 2\pi$ configuration since it can mix with the $b\,^3\Sigma^+$ Rydberg state, the parent state of the resonance. Decay to the $^1\Delta$ and $^3\Delta$ states of this configuration, although possible, is expected to be weaker, whereas resonant decay to $^1\Sigma^-$ and $^3\Sigma^-$ from the $^2\Sigma^+$ resonant state is prohibited by symmetry considerations as has been recently pointed out.[8,9]

The experimental results are consistent with the above analysis. Resonant enhancement of the $1\pi^3 5\sigma^2 2\pi$ $^3\Sigma^+$ state is expected to occur only for higher vibrational levels as a result of the large internuclear spacing of this state ($r_e = 1.352$ Å). Although it is not possible to resolve the contributions of higher vibrational levels of the $1\pi^3$-$5\sigma^2 2\pi$ configuration, there appears to be little resonant enhancement of the $^1\Delta$ and $^3\Delta$ states and no excitation, either direct or resonant, of the $^1\Sigma^-$ and $^3\Sigma^-$ states.

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