of the P_4 molecule. Thus, orbitals $2a_1'$ and $3a_1'$ of C_3H_6 strikingly resemble orbitals $4a_1$ and $5a_1$ of P_4, and there is also a relationship between the $2e'$ set of C_3H_6 and the $5t_2$ set of P_4 and between the $3e'$ set of C_3H_6 and the $6t_2$ set of P_4. These similarities are attributable to the three-atom ring structures of both molecules.

The valence electronic structure of highly symmetrical molecules exhibiting an essentially spherical shape will have the same nodal surfaces in the valence region as are to be found for an isoelectronic atom. For example, the $2a_1$ and the triply degenerate set of $1t_2$ molecular orbitals making up the filled valence shell of the methane molecule[18] exhibit the nodal structures of a 2s and a triply degenerate 2p set of atomic orbitals of neon. The tetrahedral structure of the P_4 molecule, being spherelike, should in like manner also cause the valence molecular orbitals of this molecule to exhibit pseudoatomic character. We find that orbital $4a_1$ of P_4 exhibits no nodal planes and a central concentration of charge so that it is the lowest valence s orbital of the pseudoatom. Likewise each molecular orbital of the triply degenerate $5t_2$ set has a molecular nodal plane and these three nodal planes are mutually perpendicular, so that the $5t_2$ orbitals of P_4 are pseudoatomic p orbitals. Molecular orbital $5a_1$ has a single spherical nodal surface which passes through all four phosphorus atoms. Thus from the pseudoatomic viewpoint, this is the next higher s-type pseudoatomic orbital beyond molecular orbital $4a_1$.

Orbitals $6t_2$ and 2e correspond to a fivefold set of pseudoatomic d orbitals, with each orbital of this set exhibiting the proper pair of nodal surfaces. In accord with ligand-field theory, the tetrahedral geometry of the P_4 molecule causes this fivefold set to split into a triply degenerate set of T_2 symmetry and a doubly degenerate set of E symmetry as observed. The repulsive effect of the filled-valence-shell ligands on a centrally located transition metal atom in a complex causes its t_2 set of d orbitals to exhibit a higher energy than the e set. However, for the P_4 molecule where the "ligand" positions contribute all of the electronic charge with none coming from a central atom, the situation must be reversed, with the e orbitals lying above the t_2 orbitals as observed. The calculated splitting between the t_2 and e sets, Δ_t, of 6730 cm^{-1} for the pseudoatom character of the P_4 molecule has, of course, no spectrographic significance because both the $6t_2$ and 2e levels are completely filled.

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Registry No. P_2, 12185-09-0; P_4, 12185-10-3.

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A Reinvestigation of the Optical Spectra of Nickel, Palladium, and Platinum Atoms in Noble Gas Matrices

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Optical spectra from 200 to 700 nm have been obtained for Ni, Pd, and Pt atoms in Ar, Kr, and Xe matrices at 6–8 K. The spectra for Ni and Pt atoms in Ar agree quite well with the earlier data in Ar matrices for the strongest lines. The corresponding data obtained in Kr and Xe matrices supplement the Ar data and correlate reasonably well with the reported gas-phase atomic transitions although the matrix shifts for Pt in Xe seem anomalously large. On the other hand, the previously reported optical spectrum of Pd atoms in Ar matrices, which displayed absorptions above 290 nm, is proven to have originated not from isolated Pd atoms but rather from the products of the reaction of Pd atoms with air impurities, the most intense absorptions of which are shown to be associated with $Pd(N_2)$ and $Pd(N_2)_2$ complexes. The genuine spectrum of atomic Pd in Ar, Kr, and Xe matrices is established as an extremely intense group of lines below 250 nm. The atomic transitions of Pd exhibit blue matrix shifts, in agreement with the predictions of AMCOR. An amonotonic trend is noted for the matrix-induced frequency shifts of Pd and Pt atoms in noble gas matrices ($\Delta\nu_{Xe} > \Delta\nu_{Ar} > \Delta\nu_{Kr}$) whereas a monotonic trend is seen for Ni atoms ($\Delta\nu_{Ar} > \Delta\nu_{Kr} > \Delta\nu_{Xe}$). This is taken to indicate a substantial metal–matrix interaction between the most polarizable (class B) metals and Xe, suggestive of weak "complex" formation.

Introduction

One of the simplest ways of determining the fate of a metal atom when cocondensed with an inert gas or a reactive species diluted in an inert gas is to investigate its optical spectrum. Although experimentally straightforward, the spectra can be complicated through metal atom–matrix interactions which include the shifting and broadening of spectral lines, and crystal field, spin–orbit, and multiple trapping site splitting effects. The work of Mann and Broida,[1] Brewer,[2] and Gruen[3] recently demonstrated that the observation of matrix shifts for the transitions of metal atoms isolated in noble gas matrices relative to those of the gaseous metal atom was a general phenomenon and that these shifts could be as large as several thousand wavenumbers. However, reasonable correlations between the strongest atomic transitions can be made by appropriately shifting the entire matrix spectrum from the respective gas-phase values.

Although no universal theory has been proposed to explain

Table I. Band Assignments and Matrix Shifts for Nickel ($3d^8\,4s^2$; a $^3F_6$)

Configuration	Upper state	ν_g, cm^{-1} [a]	ν_{Ar}	$\nu_{Ar} - \nu_g$	ν_{Kr}	$\nu_{Kr} - \nu_g$	ν_{Xe}	$\nu_{Xe} - \nu_g$
$3d^8\,4s\,(b^2D)\,4p$	$^3F_3$	43655	49751	6492	49261	5606	48309	4654
(a^4F)	$^3F_4$	43259			48076	4817	47169	3910
(a^4F)	$^3G_5$	43090	46511	3421	45662	2572	44642	1552
(a^4F)	$^3D_3$	42768	44444	1676	44642	2021	44052	1284
(b^2D)	$^3D_3$	42621	43668	1047	43478	857	42372	−249
(b^2D)	$^3F_4$	42585					42016	−569
(a^2F)	$^3D_3$	33501	35587	2086	34722	1221	33670	169
(a^2F)	$^3F_3$	33112	35087	1975	34246	1134	33333	221
(a^2F)	$^3G_5$	30923	33112	2189	32362	1439	31446	523
(a^2D)	$^3D_3$	29669	31545	2064	30769	1100	30030	361
(a^2D)	$^3F_4$	29481			30487	1006	29325	−156
(a^2D)	$^3F_3$	29321	30303	982	29585	264	28571	−750

[a] From ref 16.

Table II. Band Assignments and Matrix Shifts for Palladium ($4d^{10}$; $^1S_0$)

Configuration	Upper state	ν_g, cm^{-1} [a]	ν_{Ar}, cm^{-1}	$\nu_{Ar} - \nu_g$	ν_{Kr}, cm^{-1}	$\nu_{Kr} - \nu_g$	ν_{Xe}, cm^{-1}	$\nu_{Xe} - \nu_g$
$4d^9\,(^2D_{3/2})\,5p$	$^1P_1$	40839	47169	6330	46728	5889	47169	6330
					45871	5032		
$(^2D_{3/2})$	$^3D_1$	40369	45454	5085	44843	4474	45454	5085
$(^2D_{3/2})$	$^3F_2$	38811	42372	3561	42016	3205	43478	4667
					40816	2005		
$(^2D_{3/2})$	$^3P_1$	36181	40322	4141	39525	3344	42553	6372

[a] From ref 16.

these matrix shifts, simple models based on Leonard-Jones "(6-12)" and "(6-8-12)" potential functions[4] (to describe what is in essence a van der Waals interaction between the trapped atom and the matrix cage) and more recently extended Huckel calculations[5] (assuming the metal interacts with the matrix cage to form a weak complex) have been quite successful in rationalizing the gross effects of energy level shifts due to matrix perturbations.

As research interest in metal vapor cocondensation reactions continues to grow rapidly, the need for obtaining reliable, high-quality, optical data for metal atomic species in various matrix environments is becoming increasingly important. This is proving to be especially true in optical studies of matrix-controlled metal atom aggregation reactions where one wishes to identify diatomic and higher metal aggregates in the presence of isolated metal atoms.[6] Indeed, matrix atomic data are crucial to any optical study of the products of a metal vapor cocondensation reaction with a reactive species diluted in an inert matrix gas, where one wishes to distinguish the molecular spectrum of a compound from the atomic spectrum of the metal.

Recently we have reported matrix infrared and Raman spectroscopic data for the products of M/O_2,[7] M/N_2,[8] M/CO,[9] $M/N_2/O_2$,[10] and $M/N_2/CO$[11] cocondensation reactions where M = Ni, Pd, or Pt. As a continuation of these studies, we decided to investigate the optical spectra of the complexes. However, as a preliminary phase to these studies we have had to reinvestigate the optical spectra of the metals themselves, in view of the lack of available data for Ni, Pd, and Pt atoms in matrices *other* than Ar, and the distinct possibility that the spectrum previously reported for Pd in Ar[1] actually originated from impurities.

Experimental Section

Monatomic Ni and Pd vapors were generated by directly heating a 0.010 in. ribbon filament of the metal with ac in a furnace which has been described previously.[12] Platinum vapor was obtained by heating a 0.060 in. tungsten rod around the center of which were wound several turns of 0.010 in. Pt wire. The Ni, Pd, and Pt metals (99.99%) were supplied by McKay Inc., N.Y. Research grade Ar, Kr, and Xe (99.99%) were supplied by Matheson of Canada. The rate of metal atom deposition was continuously monitored and controlled using a quartz crystal microbalance.[13] The deposition rate was set such that the probability of a metal atom having another metal atom as a nearest neighbor in the matrix was approximately 1/10.[4] Matrix gas flows, controlled by a calibrated micrometer needle valve, were maintained at 2–4 mmol/hr. Typical deposition times were of the order of 30 min. The matrices were deposited onto a NaCl window cooled to 6–8 K by an Air Products liquid helium transfer system. The temperature was monitored by a thermocouple embedded into the NaCl plate. Uv–visible spectra were recorded on a Pye-Unicam SP 8000 double-beam spectrophotometer in the range 190–700 mn with an external Perkin-Elmer 165 recorder.

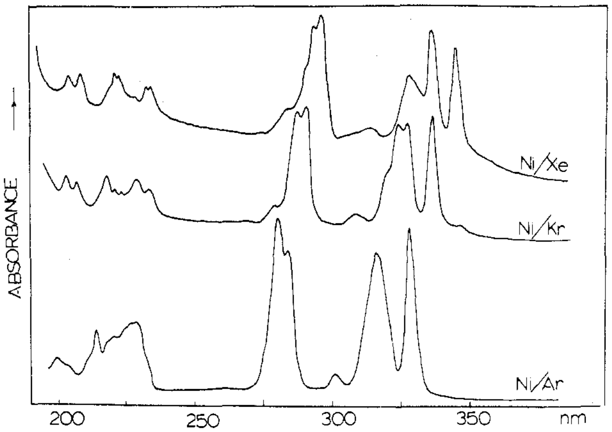

Figure 1. The optical spectra of Ni atoms isolated in Ar, Kr, and Xe matrices.

Results

Nickel. The optical spectra of nickel atoms deposited in Ar, Kr, and Xe matrices at 6–8 K are shown in Figure 1 and Table I. The spectrum in Ar is essentially the same as that reported by Mann and Broida[1] who obtained only the stronger lines. The spectra correlate reasonably well with the respective gas-phase spectra,[16] yielding matrix-induced frequency shifts which generally follow the order Ar > Kr > Xe.

Palladium. The spectra of Pd atoms isolated in Ar, Kr, and Xe matrices are shown in Figure 2 and are listed in Table II. The spectrum centers in the high-energy region below 250 nm and the observed transitions correlate reasonably well with the gas-phase spectrum.[16] Relatively large blue matrix-induced frequency shifts are observed for all lines (in the range 4400–7300 cm^{-1}) and for all three matrices. This is not unusual for high-energy transitions.[14,15] Particularly noteworthy are the surprisingly large blue shifts for Pd in Xe, which result in the amonotonic order of the matrix-induced shifts Xe > Ar > Kr (discussed later).

Table III. Band Assignments and Matrix Shifts for Platinum ($5d^9\ 6s^1$; $^3D_3$)

Configuration	Upper state[a]	ν_g, cm^{-1}	ν_{Ar}, cm^{-1}	$\nu_{Ar} - \nu_g$	ν_{Kr}, cm^{-1}	$\nu_{Kr} - \nu_g$	ν_{Xe}, cm^{-1}	$\nu_{Xe} - \nu_g$
$5d^8\ 6s^1\ 6p^1$		48779	52083	3304	51282	2503		
$5d^9\ 6s^0\ 6p^1$		46622	50251	3589	49751	3129		
					49019	2397		
$5d^8\ 6s^1\ 6p^1$		40970 40516 40194	43859	3343	43103	2587	49019	8503
$5d^9\ 6p^1$		37769 37591 37342	40983	3393	39840	2249	46511	8920
$5d^8\ 6s^1\ 6p^1$		35322	38022	2700	37453	2131	44444	6422
$5d^9\ 6p^1$		34122	36496	2374	35842	1720	41493	7371
$5d^9\ 6p^1$		32620	34722	2102	34129	1509	37313	4693

[a] Upper state undesignated in ref 16.

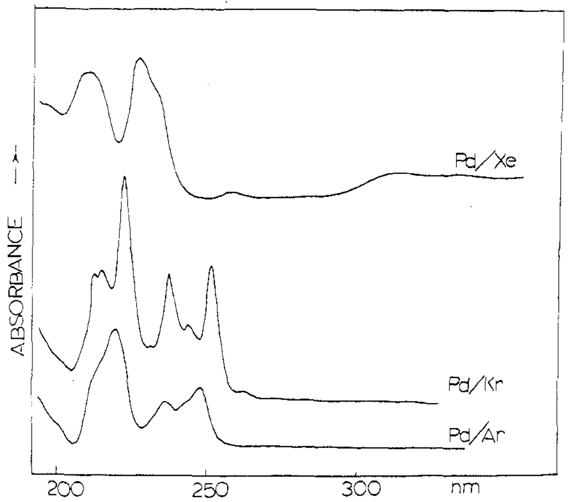

Figure 2. The optical spectra of Pd atoms isolated in Ar, Kr, and Xe matrices.

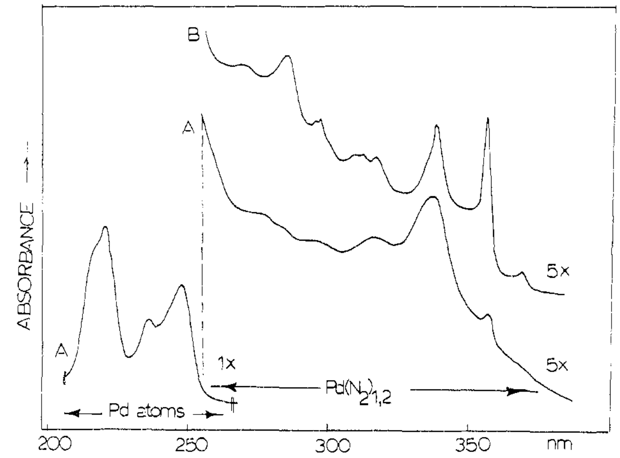

Figure 3. The optical spectrum of the products of the cocondensation reaction of Pd atoms and $^{14}N_2/Ar \simeq 1/250$ at 6-8 K, (A) on deposition, and (B) after warm-up to 20-25 K, showing the presence of Pd atoms, $Pd(N_2)$, and $Pd(N_2)_2$.

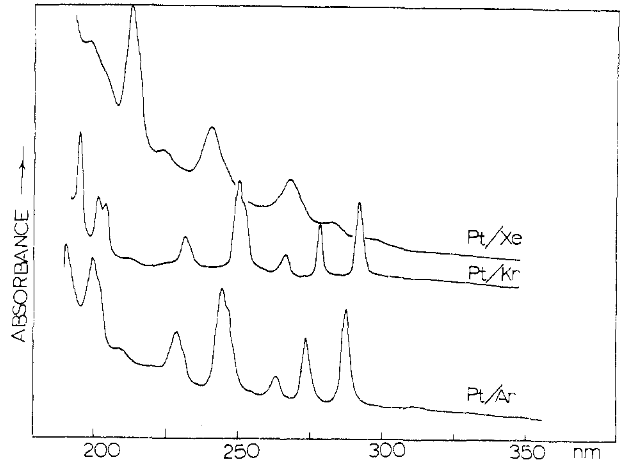

Figure 4. The optical spectra of Pt atoms isolated in Ar, Kr, and Xe matrices.

In view of our results for atomic Pd, a serious discrepancy that presently exists in the literature deserves clarification.

Mann and Broida[1] reported the atomic spectrum of Pd in Ar as consisting of four absorptions at 29600, 31260, 31940, and 33680 cm^{-1}. This was thought at the time to be an odd result as the data implied surprisingly large *red* shifts from the respective gas-phase values of the order of 6500 cm^{-1}. Red shifts of this order of magnitude have not been previously observed.

Gruen[14] recently suggested that AMCOR (atom–matrix correlations) predict the Pd atomic spectrum to occur at energies higher than 36000 cm^{-1}. This is in agreement with our Pd data, which show absorptions in the region 40000–47000 cm^{-1} for Ar, Kr, and Xe matrices. The question then arises, how does one explain Mann and Broida's data for Pd?

When Pd atoms are deposited into a $^{14}N_2/Ar \simeq 1/250$ matrix at 6°K, the major product is $Pd(N_2)$, as seen from its infrared absorptions at 2213 and 378 cm^{-1}.[8] The corresponding uv–visible spectrum is shown in Figure 3A and exhibits absorptions at 29585, 31746, and 33570 cm^{-1} which, within experimental error, correlate remarkably well with Mann and Broida's data for supposedly Pd atoms in Ar.[1] Besides the relatively weak molecular spectrum of PdN_2 in $^{14}N_2/Ar \simeq 1/250$ matrices, we observe a very intense spectrum of atomic Pd below 250 nm which passed undetected in Mann and Broida's study, as this region of the optical spectrum is fraught with recording difficulties originating from matrix scattering effects and steeply rising baselines.

It is interesting to note that on warming the matrix containing $Pd(N_2)$ to 20–25°K, $Pd(N_2)_2$ can be observed to grow in (Figure 3) as evidenced by the absorptions at 27320 and 34965 cm^{-1} in the uv spectrum and 2267/339 cm^{-1} in the infrared spectrum.[8] Similar experiments involving $Pd/O_2/Ar$ cocondensation reactions produced exceedingly weak absorptions, the implication being that Mann and Broida's optical spectra arose principally from palladium dinitrogen complexes $Pd(N_2)_{1 or 2}$ and not palladium dioxygen complexes $Pd(O_2)_{1 or 2}$.

Our experiences indicate that the most likely source of contaminants is from chemisorbed impurities on the evaporated metal films within the vacuum furnace. We found that careful outgassing of the metal filaments and furnace prior to cooling the optical sample plate was sufficient to circumvent contamination problems.

Platinum. The spectra of Pt atoms in Ar, Kr, and Xe are shown in Figure 4 and Table III. Our data for Pt in Ar are essentially the same as those previously reported by Gruen.[15]

The data in Kr and Xe are new. As with Pd, the atomic transitions for Pt in Ar and Kr can be correlated reasonably well with the gas-phase transitions.[16] However, anomalous behavior is observed in Xe, resulting in an amonotonic order of matrix-induced frequency shifts, Xe > Ar > Kr, analogous to the situation described for Pd.

Discussion

It would appear to be generally accepted that the changes in the energy levels of the various atomic states of matrix-isolated atoms result from weak interactions between the metal atom and its surrounding matrix cage. Whether the actual system is described in terms of van der Waals interactions or weak complex formation would seem to be a matter of semantics. Whatever the case, matrix interactions will induce frequency shifts away from the respective gas-phase values and will generally be different for different matrix supports. Moreover, the various atomic states of the metal will experience different perturbations from the electronic wave functions of the surrounding matrix cage which will be responsible for the different shifts observed for different atomic transitions. In this context the amonotonic trends observed for the matrix-induced frequency shifts for Pd and Pt in noble gas matrices

$$\mathrm{Xe} > \mathrm{Ar} > \mathrm{Kr}$$

compared to the more normal, monotonic trend for Ni

$$\mathrm{Ar} > \mathrm{Kr} > \mathrm{Xe}$$

could be taken to indicate a substantial metal atom–matrix interaction. This would be expected to be most pronounced for the most highly polarizable metals, Pd and Pt (soft, in the Pearson notation), with the most highly polarizable matrix material Xe. One is tempted to speculate that Pd and Pt can, in a qualitative sense, be considered to be forming a "weak complex" with Xe (i.e., a specific guest-host interaction). To confirm this proposal further experimental and theoretical studies will be required.

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Registry No. Ni, 7440-02-0; Pd, 7440-05-3; Pt, 7440-06-4; Pd(N_2), 57031-38-6; Pd(N_2)$_2$, 40810-27-3.

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Preparation of Dealuminized Mordenite

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A two-step method is described for the dealuminization of H mordenite: the zeolite is first dealuminized in the presence of steam at 538°C and then refluxed in hydrochloric acid to solubilize the removed aluminum. Unlike previous methods which either employed concentrated mineral acids below 100°C or high-temperature heating and acid extraction, the use of steam facilities the hydrolysis and the migration of aluminum from the anionic framework to cation positions. This ion-exchange reaction serves to regulate and limit the degree of dealuminization during steaming. By repeating this two-step treatment, framework aluminum has been progressively hydrolyzed and removed to yield products having silica to alumina molal ratios above 100. These highly siliceous products retain the mordenite structure and the hydrocabon sorption capacity; however, they are hydrophobic. Their hydrophobicity indicates the absence of intracrystalline silanol groups and suggests that structural rearrangement forming new $\equiv\mathrm{Si{-}O{-}Si}\equiv$ bonds takes place during steaming.

Introduction

The removal of framework aluminum from clinoptilolite, a siliceous zeolite, by leaching with boiling hydrochloric acid was first reported by Barrer and Makki in 1964.[1] Similar treatment on mordenite was reported by Dubinin and his co-workers[2,3] and Eberly et al.[4] Dealuminized mordenite with molal $SiO_2:Al_2O_3$ ratios as high as 69 was obtained from a 9.7 $SiO_2:Al_2O_3$ mordenite after leaching at 100°C for 84 h with 11 N hydrochloric acid. However partial breakdown of the crystal lattice and amorphization of the zeolite under these conditions were noted.[3]

An alternative procedure using a combination of thermal and acid treatments was reported by Sand and his co-workers.[5,6] Nearly all of the aluminum was removed ($SiO_2:Al_2O_3 > 1200$) from a 12.5 $SiO_2:Al_2O_3$ mordenite and the mordenite crystal structure was retained. The ease with which aluminum was removed by acid leaching after heating the sample to 700°C and holding there for 2 h in a dry nitrogen purge suggests that at least part of the framework aluminum was expelled from the structure during the thermal treatment. However, possibly because the procedure does not distinguish

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