lines in rare gases. This tends to support suggestions[9] that the exciton lines arise from processes involving the pairwise association of rare-gas atoms, rather than from processes more directly related to the band structure of rare-gas solids. A similar insensitivity of the rare-gas molecular spectrum to environment has been observed in other condensed phases.[10] The diatomic σ orbitals derived from Xe* 6s states probably lie near the 6s one-electron energy of ~ − 3.7 eV.[11] As this falls in the range of alkali metal band energies, it seems likely that the excited molecular orbital is virtual, rather than fully bound, in alkali metal hosts.

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Metal-Insulator Transition in Metal—Rare-Gas Alloys*

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We present the results of electrical resistivity and optical absorption measurements for compositions spanning the metal-insulator transition in RbKr and CsXe alloys. The two transitions are similar and exhibit an apparently percolative character. No marked emergence of excitons accompanies the disappearance of conductivity. Spectral features associated with metallic conduction also persist through the transitions.

Much recent discussion has focused on the nature and possible existence of Mott-type metal-to-insulator transitions in which the electron gas freezes into a nonconducting state.[1] Although no physical systems actually undergoing Mott transitions have been clearly identified, it has been suggested in several studies of rare-gas–metal mixtures[2-4] that the metal-to-insulator transition that occurs with increasing rare-gas concentration in these materials is of the Mott type. In this paper we present definitive results for the RbKr and CsXe systems. These exhibit gradual, rather than sharp, transitions to the insulating phase that can be described quite well by percolation theory. Opitcal absorption in these materials undergoes no discontinuous or even rapid variation in passing from insulating to metallic samples. Indeed "metallic" features in an energy range demonstrably associated with the metal valence electrons persist in modified form even in alloys containing 97 at.% rare gas. Similarly, "excitonic" features naturally associated with insulating hosts emerge in *metallic* hosts containing only a few percent of rare-gas atoms. The loss of conductivity observed at high rare-gas concentrations therefore appears to have no relationship to the phase transition visualized by Mott.

Figure 1 shows as open circles the resistivities at 7°K of CsXe thin-film alloys 350 to 2000 Å thick, and shows as full circles similar results for RbKr alloys. The former data are

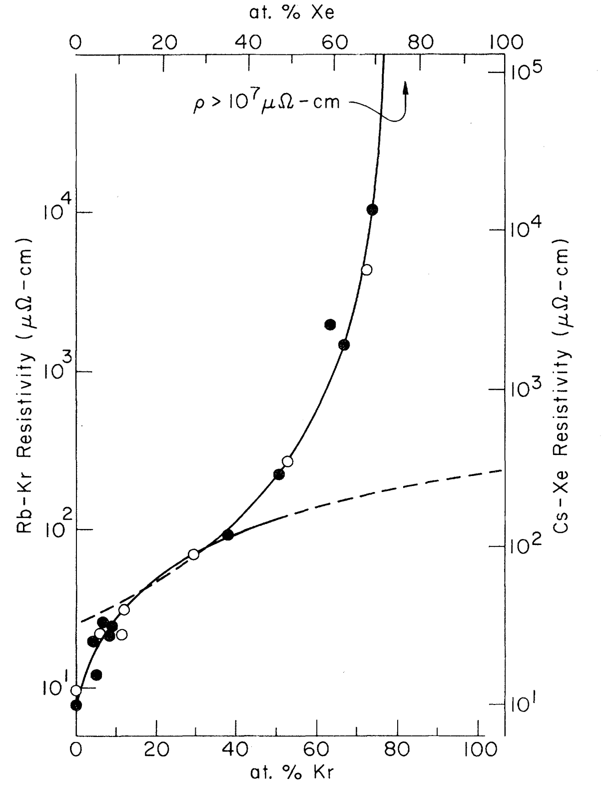

FIG. 1. The resistivities of RbKr alloys (full circles) and CsXe alloys (open circles). The full lines and their broken extrapolations represent theoretical asymptotes. Note that the scales on the RbKr axes differ from those for CsXe.

shown displaced by a factor 0.78 in resistance and 1.07 in concentration in order to display clearly the very similar trends of resistance variation with composition observed in the two systems. The samples were made by codeposition of metal and rare gas onto a He-temperature substrate from carefully crystal-calibrated sources; the rare-gas concentrations are believed to contain no more than ~7% systematic and 3% random fractional uncertainties. Film thicknesses for resistivity calculations were obtained using Vegard's law, but the huge resistivity variation is insensitive to this approximation.

It is important to establish first that the results pertain to random alloys rather than segregated samples. This is easily done for metal-rich alloys. Optical spectra presented in the preceding paper[5] show that rare-gas atoms in dilute alloys have absorption spectra quite distinct from that of the rare-gas solid. The rare-gas atoms are therefore dispersed, rather than segregated, in the host lattice. Moreover, the impurity absorption threshold agrees to ~2% with the theoretical predictions for rare-gas atoms *dispersed in the host metal.*[6] Further evidence is adduced by the resistivity results of Fig. 1. The solid line through the data at $0 < c < 30$ at.% rare gas, and its broken-line extrapolation to $c \sim 100$ at.% rare gas, indicates the expected linear dependence of residual resistivity on composition in the dilute alloys. Clearly, the linear relationship holds out to unusually large impurity concentrations, and each added rare-gas atom in the dilute alloys contributes independently to the resistance, as expected in random, but not segregated, samples. The established concentration dependence of the "pair" spectra[5] also fits perfectly with this interpretation. Finally, in confirmation of our earlier contention that the electron gas near the ground state of rare-gas impurities resembles that near vacancies,[6] we find from our results residual resistivities of 2.2 ± 0.2 and 2.8 ± 0.5 $\mu\Omega$ cm/at.% for Kr in Rb and Xe in Cs, respectively, while appropriate values for vacancies, obtained by scaling the observed values[7] of 1.5 ± 0.3 $\mu\Omega$ cm/at.% for vacancies in Au, are 2.6 and 2.8 $\mu\Omega$ cm/at.% in the two cases. The observed variation of resistance with composition is therefore precisely that expected for independent rare-gas atoms, and the existence of random short-range order in our samples appears to be established beyond reasonable doubt.

At concentrations $c \gtrsim 30$ at.% rare gas, the resistivity shown in Fig. 1 clearly exceeds the linear trend established for small c and rises rapidly to the limiting apparatus sensitivity (~12 MΩ) at a critical composition c_t of about 78 at.% Kr in Rb and ~73 at.% Xe in Cs. Using a packing factor of 0.45, appropriate for hard-sphere liquids, and hard-sphere atomic radii for liquids of the separate components, we find that the critical volume fractions of conducting material are 0.14 and 0.17 for the two systems. The percolation studies of Scher and Zallen[8] led to the value 0.15 for this parameter. Furthermore, the curve passing near the points indicates the functional form

$$\rho(c) \propto (c - c_t)^{-s}, \qquad (1)$$

with $s = 2.1$ and the critical compositions c_t indicated above. We have not been able to determine the exponent with great precision. Curves with values in the range $s = 2.0 \pm 0.5$ provide adequate fits to the data. A resistivity variation of the type given in Eq. (1) with $s \simeq 1.6$ has been ob-

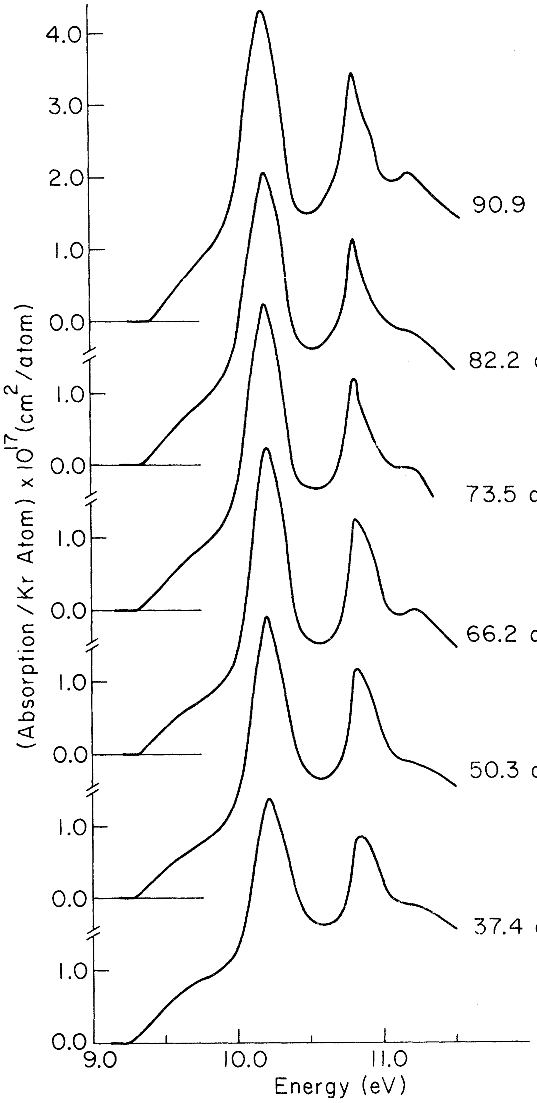

FIG. 2. Opitcal absorption in RbKr alloys. The metal-insulator transition at $c \simeq 78$ at.% Kr cannot be detected in the absorption.

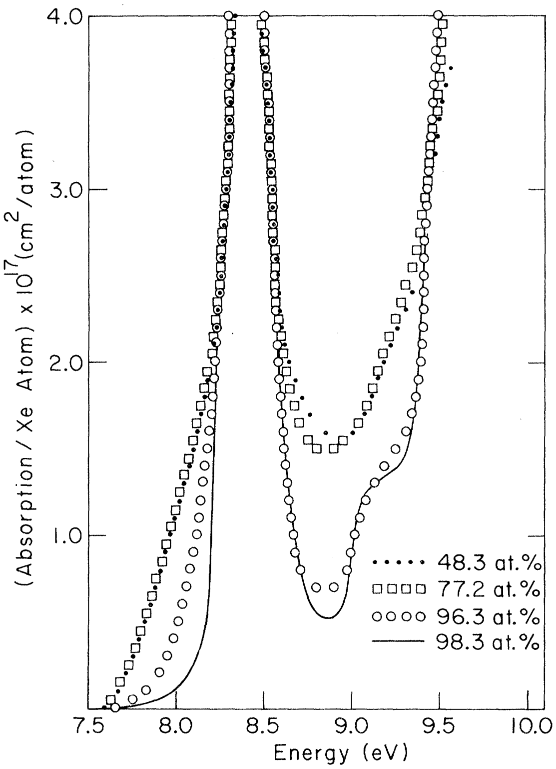

FIG. 3. Optical absorption in Xe-rich CsXe alloys, showing the threshold associated with Cs atoms finally disappearing at $c \sim 98$ at.% Xe.

tained in computer studies of percolative conduction in random networks.[9] In contrast, the Mott-type transition is believed to cause an abrupt loss of conductivity at a particular critical composition.

Any residual belief that these transitions originate in Mott-type processes appears to be completely eliminated by the optical evidence. Figure 2 shows the optical absorption *per Kr atom* for various RbKr alloys. The pair lines reported in the preceding paper[5] are detectable in RbKr alloys containing 2 at.% Kr and grow smoothly at high Kr content into the Kr exciton lines first observed by Baldini.[10] The unambiguous identification of these lines with rare-gas pair bonds is explained in Ref. 5. The "dielectric criterion" for the emergence of exciton lines, suggested by Raz *et al.*[4] for XeHg mixtures, appears to have no significance whatever in the present context. On the contrary, the strength of the excitonlike structures merely reflects the density of rare-gas pair bonds. The final feature having a critical importance concerns the broad absorption near threshold in the energy range clearly associated in metal-rich alloys with coupling between the excited electron and the host valence levels. The important point is that this absorption persists on the insulating side of the transition and exhibits no abrupt changes between alloys spanning the transition. Indeed, traces of "metallic"-type impurity absorption remain at rare-gas concentrations exceeding 90 at.%. Figure 3 shows how the "metallic" threshold features finally disappear with decreasing metal composition in CsXe alloys containing ~98 at.% rare gas.

From the present studies of RbKr and CsXe we find, in alloys possessing random short-range order, no evidence that points to an abrupt change in electronic structure at the metal-insulator transition. In fact the conduction processes appear to fall within the description of percolation theory. In a broader context the present results cast grave doubts on theories currently employed to describe the coupling between local optical excitations and the conduction electrons of a metallic host. This coupling, whose character near E_F assumes a pivotal role in determining the *theoretical* threshold profile,[11] appears much less important in the present *experimental* studies. Whereas translational invariance and metallic properties of the host are central to the theoretical models, it appears in practice that the local short-range ordering of atoms has the major influence on the optical excitation spectra of real alloys. Future theories that reflect these differing emphases will provide a more fundamental understanding of optical processes in conducting solids.

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Ferromagnetic Transition in V-Fe

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Low-field magnetization measurements on V-Fe alloys (28.5 and 29.5 at.% Fe) reveal a complex magnetic behavior of these alloys. However, the reversible part of the paramagnetic susceptibility indicates a well-defined ferromagnetic transition. Below the transition temperature, T_c, activation processes at very low fields cause the initial susceptibility to decrease rapidly as the temperature is lowered below T_c, thus explaining why the ac susceptibility displays a sharp peak at T_c.

The onset of long-range ferromagnetism in disordered V-Fe alloys is attributed to giant spin clusters.[1,2] When Fe is added in dilute amounts to V no localized moments are observed.[1,2] With increasing Fe concentration, because of statistical reasons, more and more Fe atoms are located in highly Fe-rich atomic environments. It is assumed that these Fe-rich regions can polarize spontaneously, forming giant spin clusters.[1,2] It is the interaction between these giant clusters which can lead to the onset of long-range ferromagnetism at higher Fe concentration.

There are several alloy systems in which magnetic properties are presently understood in terms of giant spin clusters, the most extensively studied being Ni-Cu.[3] Like all of these alloys, V-Fe exhibits large specific-heat anomalies for concentrations near the onset of magnetic order.[4-6] It is assumed that these anomalies are due to a local anisotropy of the giant moments, often approximated by an effective magnetic field, giving rise to a splitting of the Zeeman levels of the giant spin moments.[3,5-7]

The exponential decrease of the specific-heat