Atomic Arrangements in Gold-Nickel Solid Solutions*

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MEASUREMENTS of diffuse x-ray scattering from gold-nickel solid solutions have given results which differ markedly from the predictions of the simple statistical theory. This system shows a continuous solid solubility in a face-centered cubic lattice above 840°C; below this temperature two other face-centered cubic lattices, one rich in nickel and the other in gold, coexist. Above 840°C, therefore, the solid solution is expected to contain nickel-rich and gold-rich clusters which act as nuclei for the subsequent precipitation.

Mr. L. Seigle, of this laboratory, has shown by means of thermodynamic measurements that this system exhibits positive deviations from Raoult's Law and has positive heats of mixing. This is also interpreted as an indication that the equilibrium arrangement for the solid solution above 840°C is one which exhibits a preference for Ni-Ni and Au-Au nearest neighbor pairs. In contrast to this, the copper-gold system, above the critical temperature, exhibits positive deviations from Raoult's Law and has negative heats of mixing. X-ray measurements have shown that the equilibrium atomic arrangement in this system is one in which Cu-Au pairs are preferred.

The diffuse scattering expected from a nickel-gold solid solution containing clusters has been calculated. Such a powder pattern should show an intense small angle scattering which falls smoothly to a minimum and exhibits a weak secondary peak in the vicinity of the first lattice reflection.

Measurements of diffuse x-ray scattering from nickel-gold solid solutions have been made by both photographic and Geiger-counter spectrometer techniques. Results have been obtained for alloys containing 10 percent to 90 percent gold, and measurements have been made at 900°C as well as on specimens quenched from 900°C to room temperature. The general form of the observed scattering curve does not change on quenching and is similar for all of the compositions studied. A microphotometer trace of a typical diffraction photograph is shown in Fig. 1(a). This is a transmission powder photogram from a thin foil of Ni_3Au_2 quenched from 900°C and made in a vacuum camera with monochromatic Co $K\alpha$-radiation. The diffuse peak, which occurs at approximately $\theta = 17°$, is clearly visible. For comparison, a microphotometer trace from a similar powder photogram of Cu_3Au is shown in Fig. 1(b). The short-range order peak in the latter film occurs at a much smaller diffraction angle.

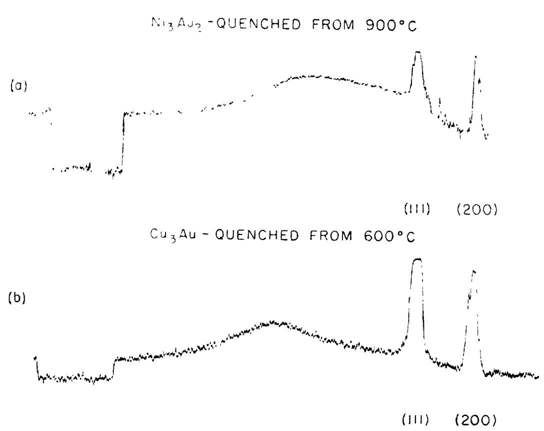

Fig. 1. Microphotometer traces of photograms of Cu_3Au and Ni_3Au_2.

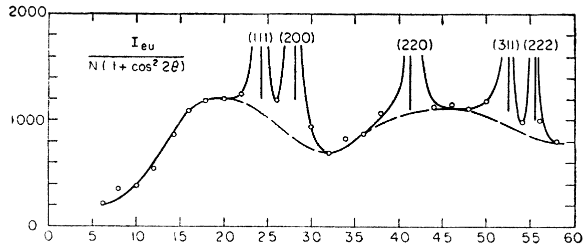

Fig. 2. Diffuse scattering in electron units from Ni_3Au_2 quenched from 900°C.

Quantitative measurements of the diffuse scattering have been made by means of a Geiger-counter spectrometer. Figure 2 shows the results for a sample of Ni_3Au_2 quenched from 900°C; the intensity in electron units was obtained by standardization with the scattering from paraffin. The form of the scattering is beyond the range of the photogram.

The diffuse scattering for Ni_3Au_2 differs considerably from that predicted by the clustering hypothesis, since there is no evidence of small angle scattering. On the other hand, the intensity distribution does not fit the form required for short range order of the Cu_3Au type. Attempts have been made by means of direct synthesis and by the use of the fourier transform to describe the results in terms of local order coefficients. Such a fit has not been possible.

These results indicate that there is no clustering of like atoms in these solid solutions. The exact atomic arrangement, however, will await further interpretation of the diffuse x-ray scattering, and further experiments with single crystals of the alloys.

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Double Beta-Decay as a First-Order Process*

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THERE are two types of double beta theories which use Fermi hamiltonians. In one, no neutrinos are actually emitted, and half-lives of the order of 10^{15} years are obtained for $3mc^2$ available kinetic energy.[1-4] In the other, two neutrinos are emitted, and the half-life is 10^{24} years for $3mc^2$ available kinetic energy.[3,5] By analyses for the products of double beta-transitions, lower limits of about 10^{19} years were found for the half-lives[6,7] of Te^{128} and U^{238}, while a positive result of 10^{21} years was found for the half-life[8] of Te^{130}. If the last datum is correct, the two-neutrino hypothesis is untenable. Also, it may be questioned whether the no-neutrino picture is correct, for the transitions examined would all have to be forbidden. Therefore, it may be interesting to consider other approaches. Of course, the data are not sufficiently good to force the adoption of a new theory. In particular, the work of Fireman[3,4] is still unexplained. Also, it is to be expected that the transitions are somewhat forbidden, for the nuclei considered are far from being mirror nuclei.

To the Fermi hamiltonian will be added a term which couples two electrons to the nucleus in the same manner as that used in Fermi theory to couple one electron and one neutrino to the nucleus. This term is to contain a bilinear combination of creation operators which create two electrons in antisymmetric states only. Let this term be

$$2^{-\frac{1}{2}}g'\{(\Psi^\dagger\Omega_e\delta\Psi^*)\Omega_N Q+\text{complex conjugate}\}.$$

g' is a constant to be evaluated from experiment. Q is Q_1Q_2, an operator which changes two neutrons into two protons. Ω_e and Ω_N are both linear combinations of the five operators occurring in beta-theory; Ω_e operates on the electron wave functions while Ω_N operates on the nuclear wave functions. Ω_N is to be interpreted as $\Omega_{N1}+\Omega_{N2}$. δ is a matrix making $\delta\Psi^*$ a spinor. Writing

$$\Psi=\sum_m a_m\psi_m$$

and using the Jordan-Wigner anticommutation relation

$$a_m^*a_n^*+a_n^*a_m^*=0,$$

one obtains, as the perturbing hamiltonian for the creation of one electron into state m and another into state n,

$$2^{-\frac{1}{2}}g'\{(\psi_m^\dagger\Omega_e\delta\psi_n^*-\psi_n^\dagger\Omega_e\delta\psi_m^*)\Omega_NQ\}.$$

For the tensor and pseudovector interactions,

$$\psi_m^\dagger\Omega_e\delta\psi_n^*-\psi_n^\dagger\Omega_e\delta\psi_m^*=0$$

while for the scalar, vector, and pseudoscalar interactions,

$$\psi_m^\dagger\Omega_e\delta\psi_n^*-\psi_n^\dagger\Omega_e\delta\psi_m^*=2\psi_m^\dagger\Omega_e\delta\psi_n^*.$$

For these last three interactions, in the "allowed" approximation for plane wave electrons, the transition probabilities are:

Scalar: $w_S(\epsilon_m,\theta)d\epsilon_m d\theta$

$$=(\epsilon_m\epsilon_n+1)\left\{1-\frac{\alpha\cos\theta}{\epsilon_m\epsilon_n+1}\right\}F.$$

Vector: $w_V(\epsilon_m,\theta)d\epsilon_m d\theta$

$$=(\epsilon_m\epsilon_n+1)\left\{1+\frac{\alpha\cos\theta}{\epsilon_m\epsilon_n+1}\right\}F.$$

Pseudoscalar: $w_{PS}(\epsilon_m,\theta)d\epsilon_m d\theta$

$$=(\epsilon_m\epsilon_n-1)\left\{1-\frac{\alpha\cos\theta}{\epsilon_m\epsilon_n-1}\right\}F.$$

Here

$$F=\frac{4\pi^3(mc^2)^5}{c^6\hbar^7}g'^2\alpha\sin\theta\frac{\epsilon_m\epsilon_n}{(1+\epsilon_m)(1+\epsilon_n)}d\epsilon_m d\theta,$$

$$\alpha=(\epsilon_m^2-1)^{\frac{1}{2}}(\epsilon_n^2-1)^{\frac{1}{2}},\quad \epsilon_n=\epsilon_0-\epsilon_m.$$

ϵ_m is the energy of an electron in the state m, and ϵ_0 is the nuclear mass difference, both in units of mc^2. θ is the angle between the electrons.

Integrating over ϵ_m and assuming that $\epsilon_0=5$, $|H^2|=1$, and the half-life$=10^{21}$ years, one finds that

$$G'\equiv g'(1/mc^2)(mc/\hbar)^3=10^{-24}.$$

It is interesting that the Fermi G is 10^{-11} for the lightest mirror nuclei; it may not be inconsistent to say that G' is G^2.

A fair approximation for the half-life as a function of ϵ_0 for $\epsilon_0>3$ is

$$\tau=2\times10^{24}\text{ years}/[\epsilon_0^2(\epsilon_0^2-4)(\epsilon_0-2)].$$

The term considered here will also make possible the emission of two electrons followed by the absorption of one of these electrons with the emission of a neutrino—that is, a second-order contribution to single beta-decay. However, in the integration over the momentum of the virtual electron, the integrand goes to zero rapidly when the wavelength of the electron becomes of the order of the nuclear diameter. It can then be seen that this contribution to single beta-decay is perhaps 10^{-30} as large as the first-order process.

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The Interpretation of Isomeric Transitions

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THEORETICAL calculations.—The half-life of nuclear isomeric states as a function of gamma-ray energy, nuclear radius, and type and multipole order of radiation has been calculated. As a simple model, it is assumed that the radiation is due to a single proton moving in the potential well of the rest of the nucleus. Radial integrals which appear are replaced by the proper power of the nuclear radius R. Because of cancellation of nuclear wave functions, this is expected to give a considerable overestimate of the values of the radial integrals and thus an underestimate of the half-life.

The half-life τ_γ for change of nuclear spin I from $L+\frac{1}{2}$ to $\frac{1}{2}$ is calculated to be

$$\tau_\gamma{}^{EL}=0.693\left[\frac{2(L+1)}{L(1\cdot3\cdot5\cdots2L+1)^2}\frac{e^2}{\hbar c}\omega\left(\frac{\omega}{c}R\right)^{2L}\right]^{-1},$$

for electric 2^L-pole radiation (EL), (1)

$$\tau_\gamma{}^{ML}=\tau_\gamma{}^{EL}\left[\frac{\hbar}{mcR}\left(\mu L-\frac{L}{L+1}\right)\right]^{-2},$$

for magnetic 2^L-pole radiation (ML). (2)

This is the theoretical half-life for gamma-emission[1] not including the effect of internal conversion. Here μ denotes magnetic moment due to intrinsic spin. The angular frequency of the radiation is ω. The mass of the proton is m. Equations (1) and (2) are nearly unchanged for any other transition for which the nuclear spin decreases by L; if the spin increases by L, the above half-lives are multiplied by the factor $(2I_{\text{initial}}+1)/(2I_{\text{final}}+1)$. Figure 1 shows both the theoretical estimates and the experimental values of the half-life of odd-A nuclei, the latter corrected for internal

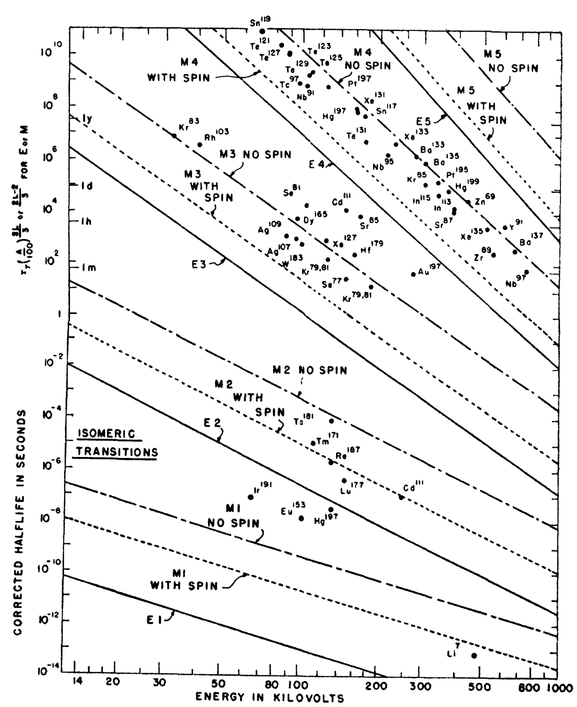

FIG. 1. Corrected half-life of odd-A isomeric transitions as function of energy. The points show experimental values of half-lives corrected for internal conversion (using theoretical values), and to $A=100$. The lines indicate theoretical predictions from the single-proton model, according to Eqs. (1) and (2), for $A=100$. (Nuclear radius, $R=1.5\times10^{-13}A^{\frac{1}{3}}$ cm.)