

ISOMERIC SHIFTS OF Sn^{119} NUCLEI IN ELECTRON COMPOUNDS OF THE COPPER-TIN SYSTEM

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Isomeric shifts on Sn^{119} nuclei in electron compounds of the copper-tin system are measured. A nonlinear dependence of the magnitude of the isomeric shift on tin concentration is observed, which evidently can be ascribed to a change in the nature of the interatomic binding forces.

THE isomer (chemical) shift of a resonance line, determined with the aid of the Mossbauer effect, characterizes the density of the s-electrons on the resonance nuclei. The determination of the isomer shifts was used in several investigations to study the redistribution of the valence electrons during formation of chemical compounds and alloying of metals. Great interest attaches, in particular, to the possibility of investigating with the aid of the isomer shift, in a sufficiently wide concentration range, the variation of the wave functions of the valence electrons of either or both components that are fused together.

Thus, the magnetic behavior of alloys of transition metals (palladium, platinum, nickel) with noble metals (such as gold) is explained on the basis of the assumption that the s- and d-bands of the transition metal overlap and its d-band is filled by valence electrons of the noble metal. At a definite concentration of the latter, at which the d-band of the transition metal is filled, the magnetic, thermal, and electric properties of the alloy change noticeably.

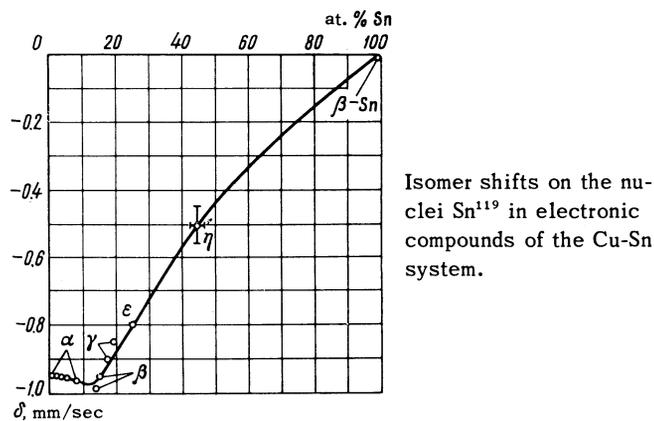
Measurements of the isomer shift in the alloy systems Au-Pd, Au-Ni and Au-Pt^[1] carried out with Au^{197} nuclei have shown no singularities in the region of concentrations corresponding to the filling of the d-band of the transition metal. This apparently can be attributed to the large degree of localization of the wave function of the d-electrons of the transition metals. Hence, to investigate the variations of the electronic properties of alloys with the aid of the isomer shift, the most favorable case would be the one in which a change takes place in the properties of the conduction band, where the wave functions of the electrons are known for the entire crystal.

The Cu-Sn system is one of those whose α , β , γ and ϵ phases can be explained in the approximation of the almost-free electrons (Hume-Rothery phases). It is therefore of interest to trace from the same point of view, with the aid of the isomer shift, the process of filling of the conduction band by the valence electrons of tin.

The alloys were prepared from components of high purity (impurity content not higher than $5 \times 10^{-4}\%$) in an argon atmosphere, with subsequent quenching in the case of the α , β and γ phases. The samples were subjects of x-ray analysis. The source of the resonance γ -radiation was magnesium stannide. The source and the absorber were at liquid-nitrogen temperature. The phases α , β , γ , ϵ and η were investigated. The absorption spectra were symmetrical singlet lines.

The figure shows a plot of the isomer shift δ against the tin content. The values of the shift are referred to a source of metallic tin. We see that in the region of the α phase, where the copper has a face-centered cubic structure, and near the start of the β phase the isomer shift decreases somewhat, while in the region of the β phase it begins to increase linearly up to the η phase.

The small change in the isomer shift in the region of the α and β phases can be understood by taking into account the large concentrations and mobilities of conduction electrons of the copper, which has a half-filled collectivized s-band. So long as this band is still unfilled, the concentration of conduction electrons near the tin ions, which give up their valence electrons to the common conduction band, is approximately constant. When the s-band of the copper becomes essentially filled (as evidenced by the change in the sign of the Hall constant for the β phase^[2]) the screening of the



tin ions in the alloy by the conduction electrons may become incomplete^[3] and the magnitude of the isomer shift should start to change.

The phases for which the isomer shifts are located farther on the increasing linear section of the curve, exist in a very narrow range of compositions, especially the ϵ and η phases corresponding to the compound Cu_3Sn and Cu_6Sn_5 ^[4]. From this we can assume that, starting at least with the γ phase, the influence of tin on the band structure of copper becomes quite noticeable. An even more noticeable influence can be exerted by the covalent bonds, whereas for the α and β phases the predominating bond should be of the metallic type.

The negative sign of the isomer shift for the phases (relative to the metallic tin) indicates a large filling of a number of s-states of tin in the alloy, compared with the pure tin, if account is taken of the negative sign of the change in the charge radius of Sn^{119} nucleus on going over from the first-excited to the ground state^[5,6].

Insofar as we know, in the hitherto investigated binary systems the observed connection between the isomer shift and the concentration was linear. Thus, such a relation was observed both for the Pd-Sn system, where intermetallic compounds are predominantly formed and the region of solid solutions is small^[7], and for the formation of systems Au-Pd, Au-Ni, and Au-Pt^[1], where the solid solutions exist in the entire range of concentrations. In each of these systems, the nature of the chemical bond remained unchanged if we disregard the relatively small region of existence of solid solutions in the Pd-Sn system. The Cu-Sn system is apparently the first among the systems with a non-linear dependence of the isomer shift on the concentration.

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69