RELATION BETWEEN THE ISOMER SHIFTS FOR THE 23.8-keV GAMMA TRANSITION OF Sn¹¹⁹ IN METALLIC SOLID SOLUTIONS AND THE DYNAMICAL PROPERTIES OF THE MATRIX

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Isomer shifts have been measured for the 23.8-keV γ transition in Sn¹¹⁹ introduced as an impurity in various metallic matrices. A simple and unique connection is established between the electron density at the nucleus of the impurity atom and the dynamical characteristics of the host. A possible qualitative interpretation of this connection is discussed.

RECENTLY^[1,2] we have measured the probability for recoilless absorption of 23.8-keV γ quanta by Sn¹¹⁹ nuclei embedded as impurities in matrices of vanadium, gold, platinum and thallium. The good agreement of the results with the theory^[3] of the Mössbauer effect on impurity nuclei in monatomic lattices showed, in particular, that the results can be interpreted on the assumption that the force constants are unchanged (i.e., the force constants for the impurity in the matrix are equal or close to the force constants for the host atoms).

It is convenient to Introduce an effective force constant K, characterizing the interaction between the atoms, using the following expression for the Debye temperature of the monatomic regular crystal (the host):

$$\Theta_0 = \text{const} (K / M_0)^{\frac{1}{2}},$$
 (1)

where M_0 is the mass of the host atoms. The fact that the interaction is unchanged or is changed only slightly when an impurity is introduced into a metallic matrix (which has been established, at least, for a tin impurity atom) permits the assumption that the coupling of the impurity atom to the atoms of the host will be characterized by an effective force constant which is equal or close to the effective force constant of the matrix $K \sim \Theta_0^2 M^2$. One can then describe the probability for the Mössbauer effect on the impurity nucleus and its temperature dependence in the Debye approximation by using a model of a monatomic regular crystal with an effective Debye temperature

$$\Theta = \operatorname{const}(K / M)^{\frac{1}{2}} = \Theta_0(M_0 / M)^{\frac{1}{2}}, \tag{2}$$

where M is the mass of the impurity atom. The validity of formula (2) is illustrated in Table I by data on the probability for the Mössbauer effect on impurity Sn^{119} nuclei. The effective Debye temperature Θ were computed from the experimental values of the probability f'. (Table I gives values of f' only for temperatures of 77 and 290°K, although f' and Θ were actually determined over a wide range of temperatures.)

It was noted that for matrices with similar values of the effective force constant (and consequently with similar values for the Mössbauer probability), the isomer shifts of the 23.8-keV transition were also similar. This indicated that there is a connection between the effective force constants and the size of the isomer shift. To establish this relation we have measured the isomer shift for the 23.8 keV γ transition in Sn¹¹⁹ in various metallic matrices (Table II). The measurements were made on solid solutions with low tin concentrations (1-3 at %). The data were compared to the parameter $\Theta_0^2 M_0$, which is proportional to the effective.

Table I

Matrix	f		0.077	$\Theta \left(M/M_{0} \right)^{1/2}$	
	290° K	77° K	Θ°K	°K	e₀, °K
V Pt Au Tl Ag		$\begin{array}{c} 0.78 \pm 0.03 \\ 0.80 \pm 0.05 \\ 0.72 \pm 0.04 \\ 0.35 \pm 0.03 \\ 0.68 \pm 0.04 \end{array}$	$\begin{array}{c} 260 \pm 10 \\ 292 \pm 15 \\ 220 \pm 10 \\ 117 \pm 5 \\ 207 \pm 15 \end{array}$	$\begin{array}{c} 396 \pm 15 \\ 228 \pm 12 \\ 171 \pm 8 \\ 90 \pm 4 \\ 217 \pm 16 \end{array}$	390 220 170 96 210

Table II

Matrix	δ , mm/sec	⊖₀, °K	$\Theta_0^2 M_0 \cdot 10^{-4}$
Ag Cu Au In Tl Pb V Pt Fe Pd	$\begin{array}{c} +0.20\pm0.02\\ -0.13\pm0.03\\ +0.15\pm0.02\\ +0.96\pm0.05\\ +1.01\pm0.03\\ +1.20\pm0.05\\ -0.27\pm0.04\\ -0.42\pm0.06\\ -0.42\pm0.06\\ -0.37\pm0.03\end{array}$	$210 \\ 325 \\ 170 \\ 129 \\ 96 \\ 90 \\ 390 \\ 220 \\ 420 \\ 275$	$\begin{array}{r} 4.76 \\ 6.71 \\ 5.69 \\ 1.91 \\ 1.88 \\ 1.68 \\ 7.74 \\ 9.44 \\ 9.85 \\ 8.04 \end{array}$
Sn	$+0.69 \pm 0.03$	170	3.43

tive force constant. (The corresponding values of this parameter and the literature data on the Debye temperatures for the matrices are given in Table II. The shifts are given for a temperature of 77°K, relative to the energy of the transition in Mg₂Sn, which was used as the source. The dependence of the isomer shift δ on the parameter $\Theta_0^2 M_0$ is shown in the figure. We see that the connection between δ and $\Theta_0^2 M_0$ is surprisingly simple and definite.

Since both the isomer shift and the dynamical parameters of the lattice depend on the nature of the interatomic coupling, there should in general be a connection of some sort between δ and the dynamical parameters. But the simplicity of the relation observed in the metallic solid solutions which we have studied is unexpected, since the electron density at the nucleus is generally related in a complicated way to the quantities characterizing the interaction between the atoms. A theoretical treatment of the force constants for metals was given by Brenig, ^[4] who showed, in particular, that in first approximation the force constants are determined by the change of the electron density during displacements of the nuclei from their equilibrium positions. But the approximation used in his work can hardly be taken over to explain the observed relation for all the solid solutions studied by us.

We see from the figure that the electron density at the impurity nucleus is directly and practically uniquely related to the value of the effective force constant. An increase of the force constant corresponds to a decrease of the electron density at the impurity nucleus. An explanation of the observed relation is faced with difficulties, especially as the metals used as matrices have very different properties (ranging from monovalent metals to the transition metals). One can, however, try to give an explanation of this correlation. In doing this it is natural to start from the fact that when an impurity is introduced the force constants are unchanged (or change insignificantly). If we assume that when the tin atom is embedded



FIG. 1. Relation of isomer shift δ of the 23.8-keV γ transition in Sn¹¹⁹ in various metallic matrices to the dynamical parameter of the host $\Theta_0^2 M_0$. The isomer shifts are measured at 77°K, relative to the energy of the γ transition in Mg₂Sn.

in the metallic matrix all its valence electrons are collectivized (being put into the conduction band or used for covalent bonding), then to make the force constants equal there must be a screening of the tin ion by the conduction electrons. The stronger the interaction between the atoms of the host (and consequently between the impurity atom and the host atoms), the weaker should this screening be. In other words, to a large value of effective force constant there corresponds a large effective charge of the tin ion. If we assume that the electron density at the impurity nucleus is the sum of the density due to the ion's own electrons and the density of conduction electrons localized in the region of the nucleus, we should conclude that there is a unique connection of the isomer shift to the quantity characterizing the atomic interaction.

We mention that another argument is possible, based on the assumption that the effective number of collectivized electrons of the impurity atom is different in different matrices and is proportional to the interaction between the host atoms. It is not hard to see that this is equivalent to the previous argument so far as the present result is concerned.

In addition to the correlation between δ and $\Theta_0^2 M_0$, one also observes a relation between the isomer shift and other parameters of the host lattice. For example, there is a clear correlation between the shift δ and the cohesive energy, the number of atoms per unit volume, and the separa-

tion between nearest neighbors. One might suppose that the existence of such correlations is a consequence of the connection between these parameters and the value of the effective force constant. It is also possible that to explain them one should consider the relation of these parameters to the electron density of the host.

In conclusion we point out an interesting possibility arising in studies of the dependence of the isomer shift on the effective force constant. If one finds this dependence for the region of small K, then extrapolating the curve to K = 0 gives the value of the isomer shift for the free atom. The curve in the figure still does not permit a reliable extrapolation, but in principle a study of the dependence as discussed above may be useful for an absolute normalization of the isomer shift. The authors thank Yu. Kagan for valuable discussions of the results, and also P. L. Gruzin, in whose laboratory some of the alloys were prepared.

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