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THE MÖSSBAUER EFFECT ON Sn^{119} *NUCLEI IN A VANADIUM MATRIX*

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We have measured the probability for recoilless resonant absorption of 23.8 keV γ quanta by Sn^{119} nuclei contained in a vanadium matrix, over the temperature interval from 77 to 660°K. The measurements were made on solid solutions with a tin concentration of 1.6, 3.45, and 7.22 at.%. A direct comparison was made with the theory developed in [3]. We used the spectral density function for the vanadium phonon spectrum which was found experimentally.[11] There is good agreement between experiment and theory over the whole temperature range. Within the limits of error no differences were found for the samples with the three different concentrations.

1. INTRODUCTION

THE probability for the Mössbauer effect, its temperature dependence, and the temperature red shift of the resonance line are directly related to the phonon spectrum of the crystal. To calculate these quantities one must know the spectral density of the squared displacement of the radiating (or absorbing) nucleus along three mutually perpendicular directions depending on the vibration spectrum. But both the probability for the Mössbauer effect and the temperature shift are integral quantities in the phonon spectrum. Consequently their dependence on particular features of the spectrum is greatly reduced. In particular, this has the consequence that for the simplest case of a monatomic cubic lattice an approximate description of the effect is already given by a one-parameter description of the spectrum, the Debye model. (We might mention that the Debye temperature given in this way may differ considerably from the value given by specific heat measurements at low temperatures.)

The picture becomes much more complicated when we go to an anisotropic monatomic crystal,[1] or when we consider a crystal whose unit cell contains several atoms. It is then necessary to have more detailed information about the vibrations of the radiating (or absorbing) nucleus, and a one-parameter description of the spectrum becomes completely inadequate. Thus, when there are optical branches of the vibration spectrum, the Debye temperature ceases to be even an approximate characteristic of the phonon spectrum.

It is of particular interest to study the Mössbauer effect on impurity nuclei in a crystal, when the dependence of the effect on the vibration spectrum of the host becomes especially complicated. The theory of this phenomenon was developed by Kagan and Iosilevskiĭ[3] (cf. also [4]). Later some of the results were obtained by another method by Dzyub and Lubchenko.[5] Maradudin and Flinn[6] described the effect in the classical limit, and very recently Visscher[7] has given the results of a machine calculation for a particular simple model of a crystal.

As was shown in [3,4], the spectral density of the squared displacement for an impurity atom is very different from that for an ideal lattice. It depends essentially on the mass m' of the impurity atom, on the change in the force constants and on the spectrum of the ideal lattice. For an isolated impurity atom the local substructure of the lattice seems to give a weakening of the effect of a change in the force constants. This permits one to assume that for a large difference between the mass m' of the impurity and the mass m of the host, one can in first approximation neglect changes in the force constants. For this case a general expression was given in [3] for the Mössbauer effect on an impurity atom in a monatomic lattice for an arbitrary mass ratio m'/m and over the whole range of temperatures. When the host has cubic symmetry, the probability for the effect is given by the formula

$$f = f\{\Psi(\omega),\ m'/m,\ T\}, \qquad (1)$$

where $\Psi(\omega)$ is the frequency distribution function of the phonon spectrum. For a comparison of theory with experiment it is expedient to use a host for which the function $\Psi(\omega)$ is known. So far there has been only one case of a reliable experimental determination of $\Psi(\omega)$ for a monatomic crystal. This is vanadium, whose lattice dynamics have been investigated by incoherent scattering of slow neutrons in several experiments.[8-11] For just this reason, since we wanted a direct comparison of theory and experiment, we chose vanadium as the host and studied the Mössbauer effect on Sn^{119} nuclei introduced into this matrix. It is known that tin dissolves in vanadium, forming a solid solution for tin concentrations up to 10 at.%.[12] We therefore decided to do the experiments at several tin concentrations, in order to extrapolate the results to zero concentration, and also in order to determine the concentration at which the interaction between the impurity atoms begins to affect the magnitude of the effect.

2. DESCRIPTION OF THE EXPERIMENT

A solid solution of tin in vanadium was prepared by melting normal tin together with vanadium having a purity of 99.75%. The tin content of the solution was determined by chemical analysis. Measurements were made using as absorbers solid solutions with three different concentrations: 1.6, 3.45, and 7.22 at.% of tin. The absorbers were prepared from a fine powder of the alloy by deposition on aluminum foil. The powder was first heated in vacuum at 900°C for 90 hours; control measurements showed, however, that the annealing has no noticeable effect on the shape of the absorption spectrum or the size of the effect. Absorbers were made with several thicknesses from 3.5 to 8 mg/cm^2 (of tin); for measurements at larger thicknesses we used two or more absorbers together. The source of γ rays was tin dioxide, SnO$_2$, containing the Sn^{119m} isomer; the source thickness was 9 mg/cm^2. The source was kept at room temperature for all measurements.

The measurements of the resonance absorption spectrum were made on an apparatus in which the absorber was given a constant velocity relative to the source by means of a three-sector cam (the sectors of the cam correspond to positive, negative and zero velocity of the absorber). The rate of rotation of the cam could be varied by changing the current to an electric motor, whose rotation frequency was kept constant during the measurements by means of an electronic stabilization circuit using a tachometer generator. The detector of the γ quanta passing through the absorber was a scintillation counter with a NaI(Tl) crystal, 1.5 mm thick. For measurements below room temperature the absorber was placed in a small cryostat whose temperature could be varied smoothly by changing the current through a heater in contact with the liquid nitrogen. For measurements above room temperature the source was placed in a cylindrical furnace with its axis along the direction of motion of the quanta. The temperature was measured with a copper-constantan thermocouple; during the measurements the temperature was maintained constant to 2—3°.

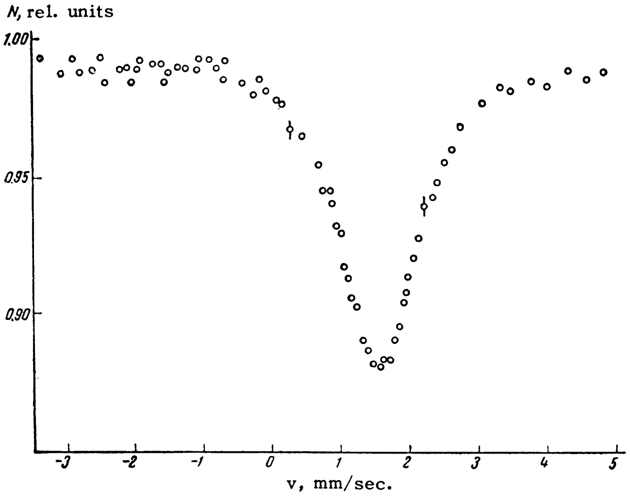

FIG. 1. Resonance absorption spectrum for a 7.8 mg/cm^2 (tin) absorber at room temperature (the abscissa is the velocity of the absorber, the ordinate gives the flux of γ quanta).

3. RESULTS

Resonance absorption spectra were taken over the temperature range from 77 to 660°K with absorbers prepared from alloys with three different concentrations of tin in vanadium (1.6, 3.45, and 7.22 at.%). We determined the effect ϵ, the line width Γ_{exp} in the absorption spectrum and the position of the line. At the two extreme temperatures (77 and 660°K) and at room temperature we made careful measurements of absorption spectra for samples of all three concentrations and for different thicknesses in the Sn^{119} isotope.

We found that within the accuracy of the measurements, at each temperature the line width and the position of the line were the same for all three tin concentrations. This somewhat unexpected result was checked on absorbers of several different thicknesses. The absence of any noticeable dependence of the resonance absorption on concentration allowed us, in measuring the temperature dependence of the effect at intermediate points, to use samples with the maximum tin concentration, which has obvious advantages from the point of view of experimental precision.

A typical resonance absorption spectrum, obtained with a 7.8 mg/cm^2 (tin) absorber at room temperature, is shown in Fig. 1. The spectrum consists of a single line; the shift relative to the line in SnO_2 is +1.560 ± 0.015 mm/sec.

To determine the absolute value of the effect, f′, we used the standard procedure of measuring the dependence of ϵ, Γ_{exp} and the area under the line as a function of absorber thickness. The experimental data for the magnitude of the effect and the line width are shown in Fig. 2. The experimental results must be compared with the theoretically calculated dependence of ϵ, Γ_{exp} and area under the line as a function of the absorber thickness C_a ($C_a \sim f'$).

To calculate the theoretical dependence one must know both the width of the emission line Γ_s and the width Γ_a of the absorption line. The width of the emission line was determined from the dependence on absorber thickness of the line width for an absorber made of SnO_2. A value of 0.67 ± 0.02 mm/sec was found for Γ_s, which is very close to the result of Pham Zuy Hien and Shpinel',[13] obtained with a source of about the same thickness. Knowing Γ_s and the temperature dependence of Γ_{exp} (Fig. 2), it is easy to determine Γ_a by extrapolation to zero thickness. Its value was 0.48 ± 0.03 mm/sec, which is somewhat larger than the natural width corresponding to the lifetime of the excited state.

Because the nature of the broadening of the line for SnO_2 has still not been completely explained, the theoretical curves were computed on two assumptions: a) the shape of the emission line is Lorentzian and b) the broadening of the emission is caused by quadrupole interaction. In particular, in making the computations we used the method for treating the spectra which was proposed in [13-15]. The experimental results were compared with the theoretical dependence of ϵ, Γ_{exp} and the area of the line on the thickness C_a. The values of C_a found in this way depended very slightly on the method used for determining them: they were very close, within the limits of error of the measurements. The quantity f′ was then determined by the usual formula $f' = C_a(\Gamma_a/\Gamma)\sigma_0 n$, where Γ is the natural width of the level, n is the number of Sn^{119} nuclei per cm^2, and

$$\sigma_0 = [(2I^*+1)/(2I_0+1)]\lambda^2/2\pi(1+\alpha)$$

(I_0 and I^* are the spins of the ground and excited states of the nucleus, λ is the wave length of the γ radiation, and α is the total internal conversion coefficient). In the computations we used the values $\Gamma = 0.31$ mm/sec, $\sigma_0 = 1.15 \times 10^{-18}$ cm^2. We mention that over the whole temperature range Γ_a was practically constant, which corresponds to a universal relation between ϵ, Γ_{exp} and the effective thickness C_a.

The values of f′ found at all temperatues are shown in Fig. 3. The precision in the absolute values of f′ is 10—12% at room temperature and lower, and reaches 12—15% at high temperatures. A considerable part of the error comes from uncertainty in the determination of Γ_a, so that the error in the determination of the relative change of f′ with temperature is much smaller.

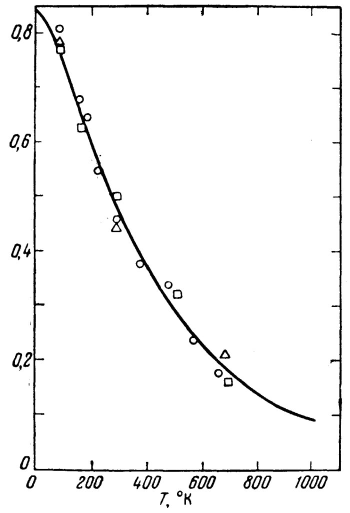

FIG. 3. Temperature dependence of probability for recoilless resonance absorption by Sn^{119} nuclei in a vanadium matrix, for different concentrations of tin in solid solution: □ – 1.6 at.%, △ – 3.45 at.%, ○ – 7.22 at.%. The solid curve is the theoretical computation.

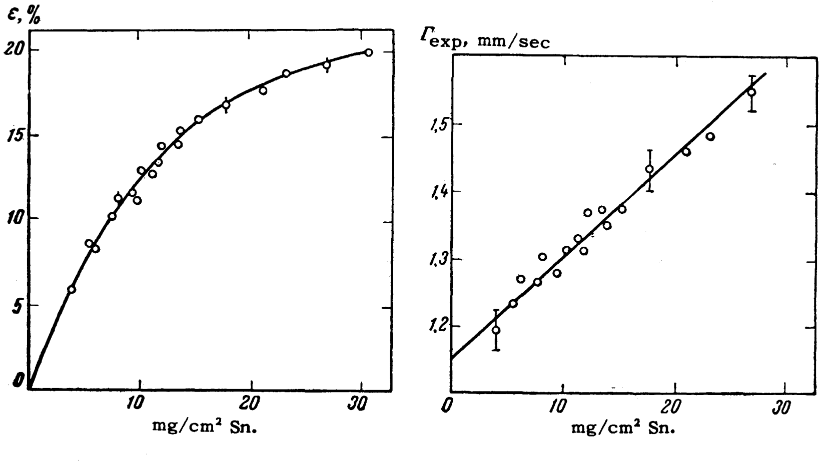

FIG. 2. The magnitude of the effect, ϵ, and line width of absorption spectrum, Γ_{exp}, as a function of absorber thickness, at room temperature (the absorber thickness in tin is given).

4. CALCULATION OF PROBABILITY OF THE EFFECT

According to the results found by one of the authors in collaboration with Iosilevskiĭ,[3] the probability for the Mössbauer effect on an impurity nucleus in a cubic crystal when $m'/m > 1$ can be written in the form

$$f' = \exp\left\{-\frac{R'(1-\varepsilon)}{\hbar}\int_0^{\omega_{0max}} d\omega\,\frac{\Psi(\omega)}{\omega}\,\frac{[2\bar{n}(\omega)+1]}{[1-\varepsilon S(\omega^2)]^2+[{}^1/{}_2\pi\varepsilon\omega\Psi(\omega)]^2}\right\},$$

$$S(\omega^2) = \omega^2 \fint_0^{\omega_{0max}} d\omega'\,\frac{\Psi(\omega')}{\omega^2-\omega'^2}\,. \qquad (2)$$

Here $R' = E_\gamma^2/2m'c^2$ is the recoil energy for the impurity nucleus, $\epsilon = (m-m')/m$, $\bar{n}(\omega) = (e^{\hbar\omega/kT}-1)^{-1}$. In the computations we used the frequency distribution function $\Psi(\omega)$ for the vanadium matrix which was found by Zemlyanov et al. [11] These results, especially when we consider the great care in treating the data, may be regarded as the most reliable available at present.¹⁾ The results of the numerical computation of (2) over the temperature interval from 0 to 1000°K are also shown in Fig. 3.

5. DISCUSSION OF RESULTS

The present results show that within the accuracy of the experiment there is good agreement between the measured and calculated values of f′ over a wide range of temperatures. In accordance with the general prediction, the effect drops much more rapidly with temperature than for an ideal lattice with the same phonon spectrum. If we use the crude one-parameter description, the temperature dependence $f'(T)$ is gotten approximately by replacing the Debye temperature Θ_D by the temperature $\Theta_D\sqrt{m/m'}$, in complete agreement with the result found in [3]. The observed agreement between the experiments and the computations from formula (2), already noted in [4] in connection with the experiments of Shirley et al [15] and Heberle et al [16], leads us to believe that the assumption of small change in the force constants is actually confirmed.

In the present work, within the experimental accuracy, no dependence of the effect on tin concentration in the vanadium host was observed over a quite wide range of concentrations. This (to us) unexpected result made it unnecessary to extrapolate to zero concentration, but the magnitude of the concentration at which the change in f′ begins to be significant remains an open question. It would be of interest to examine whether our result is related to the nature of the solid solution formed by tin and vanadium.

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[1] Yu. Kagan, DAN **140**, 794 (1961), Soviet Phys. Doklady **6**, 881 (1962).

[2] Yu. Kagan, JETP **41**, 659 (1961), Soviet Phys. JETP **14**, 472 (1962).

[3] Yu. Kagan and Ya. A. Iosilevskiĭ, JETP **42**, 259 (1962), Soviet Phys. JETP **15**, 182 (1962).

[4] Yu. Kagan and Ya. A. Iosilevskiĭ, JETP **44**, 284 (1963), Soviet Phys. JETP **17**, 195 (1963).

[5] I. P. Dzyub and A. F. Lubchenko, DAN **147**, 584 (1962), Soviet Phys. Doklady **7**, 1027 (1963).

[6] A. A. Maradudin and P. A. Flinn, Phys. Rev. **126**, 2059 (1962).

[7] W. M. Visscher, Phys. Rev. **129**, 28 (1963).

[8] A. T. Stewart and B. N. Brockhouse, Revs. Modern Phys. **30**, 250 (1958).

[9] Eisenhauer, Pelah, Hughes, and Palevsky, Phys. Rev. **109**, 1046 (1958).

[10] K. C. Turberfield and P. A. Egelstaff, Phys. Rev. **127**, 1017 (1962).

[11] Chernoplekov, Zemlyanov, and Chicherin, JETP **43**, 2080 (1962), Soviet Phys. JETP **16**, 1472 (1963). Zemlyanov, Kagan, Chernoplekov, and Chicherin, IAEA Conference, Chalk River, 1962; vol. II, p. 125.

[12] A. E. Vol, Stroenie i svoĭstva dvoĭnykh splavov (Structure and Properties of Binary Alloys), Fizmatgiz, 1962.

[13] Pham Zuy Hien and V. S. Shpinel', JETP **44**, 393 (1963), Soviet Phys. JETP **17**, 268 (1963).

[14] G. A. Bykov and Pham Zuy Hien, JETP **43**, 909 (1962), Soviet Phys. JETP **16**, 646 (1963).

[15] Shirley, Kaplan, and Axel, Phys. Rev. **123**, 816 (1961).

[16] Heberle, Parks, and Schiffer, Preprint, 1962.

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1) The results given in [11] must be divided by $\sqrt{\omega}$ in order to get the true value of $\Psi(\omega)$ (and then, of course, one must normalize the function). This step was inadvertently omitted in [11].