THE MÖSSBAUER EFFECT ON Sn-119 NUCLEI

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Abstract

Full Text

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IN TIN FILMS DEPOSITED AT 4.2°K

The study of films deposited at low temperatures is of great interest, since they possess a number of specific properties, the principal one of which is undoubtedly an elevated superconducting transition temperature. For example, tin films condensed at 4.2°K have a critical temperature of the superconducting transition of 4.3-4.6°K ([1]). We have therefore measured the resonant absorption of γ rays in tin films deposited at 4.2°K on a glass substrate.

The experimental apparatus on which the measurements of resonant absorption were carried out is described in ([2]). Radiation with an energy of 23.8 keV, arising in the decay of Sn-119m nuclei, was recorded with a scintillation counter. The Mössbauer absorption spectrum was taken point by point with the absorber moving relative to the source, for which the compound Sn^*O_2 was used. The absorber was a film of tin enriched to 88.1% in isotope 119. The film was obtained by evaporating small charges of tin from tungsten evaporators onto the inner walls of a spherical glass bulb immersed in liquid helium. The bulb had previously been degassed and annealed at 180°C in a vacuum of $2 \cdot 10^{-6}$ torr for 10-12 hours.

Fig. 1. Typical curves of superconducting transitions corresponding to amplitudes of resonant absorption:

Fig. 2

Figure 2: Fig. 2

1 $-(0.6 \pm 0.1)\%$; 2 $-(1.8 \pm 0.1)\%$; 3 $-(3.4 \pm 0.1)\%$; 4 $-(4.9 \pm 0.1)\%$. In this thickness range the dependence of the amplitude on thickness is linear, and 1% of amplitude corresponds to 125 Å.

The superconducting transition was recorded by measuring the mutual inductance of two coils rigidly fixed to the bulb. One of the coils was fed with an alternating current of frequency $\sim$ 70 kHz, corresponding to the resonant frequency of this system. The transition to the superconducting state was determined from the change in the signal taken from the second coil, which was fed to the input of a resonant amplifier. This circuit had high sensitivity, which made it possible to work at low input-signal levels. Typical curves of superconducting transitions are shown in Fig. 1. Along the ordinate is plotted the relative value of the signal taken from the second coil. The critical temperature T_k was determined from the onset of the transition and varied, for the various 20 films measured by us, from 4.2 to 4.55°K, while the widths of the transition curves ΔT_k ranged from 0.015 to 0.145°K. We note that, as a rule, thinner films had lower T_k at a larger value of the transition width.

The resonant-absorption curve was taken point by point at 4.2°K for each of the films obtained in the freshly condensed state and after–

after annealing at 290° K. For some films deposited at the helium temperature, the absorption spectrum was also recorded in the normal state, but no difference within the experimental errors was found in the absorption spectra for films in the normal and superconducting states. No difference was found either in the absorption spectra for freshly deposited tin films and films annealed at 290° K with thickness greater than $5 \cdot 10^{-6}$ cm. On the other hand, for cold-deposited films with thickness less than $3 \cdot 10^{-6}$ cm, a noticeable distortion of the absorption spectrum is observed: a shift of the maximum-absorption peak toward higher energies,

Fig. 2. Resonance-absorption curves of films in the cold-deposited state (upper curves) and in the annealed state (lower curves). *a*–measurements in the superconducting state, *b*–measurements in the normal state. In the annealed state all curves correspond to identical values of the resonance velocity $v_p = 2.55$ mm/sec and half-width $\Gamma_{1/2} = 1.4$ mm/sec. In the cold-deposited state, for film 1, $v_p = 2.75$ mm/sec, $\Gamma_{1/2} = 1.8$ mm/sec, film thickness $\Delta = 150$ Å; for film 2, $v_p = 2.75$ mm/sec, $\Gamma_{1/2} = 1.8$ mm/sec, $\Delta = 220$ Å; for film 3, $v_p = 2.80$ mm/sec, $\Gamma_{1/2} = 1.9$ mm/sec, $\Delta = 250$ Å; for film 4, $v_p = 2.75$ mm/sec, $\Gamma_{1/2} = 2.0$ mm/sec, $\Delta = 120$ Å

broadening and asymmetry of the resonance curves. After annealing at 290° K, all these distortions completely disappeared and the resonance curve did not

Fig. 3

Figure 3: Fig. 3

differ in any way from the absorption curve in a polycrystal.

Characteristic absorption curves for several films of different thicknesses after condensation and after annealing are shown in Fig. 2. The magnitude of the resonance absorption is represented by the relative absorption amplitude. During the measurements the counting rate was 1200 counts/sec and, for each point, statistics of $2 \cdot 10^6$ pulses were accumulated. Since such measurements required a long time, instead of recording the velocity spectrum in detail, absorption measurements were carried out for 10 fixed points near the resonance velocity. When recording the absorption spectrum for freshly condensed films in the normal and superconducting states, in particular, measurements were carried out continuously over 2 days.

As is seen from Fig. 2, the position of the maximum-absorption peak for freshly condensed tin films, determined by the intersection of straight lines drawn through points near the half-width of the curve, is shifted toward higher energies by (0.20 ± 0.06) mm/sec. The absorption curves are broadened and asymmetric, having a steeper slope on the higher-energy side. The broadening of the absorption line, defined as the difference between the half-width of the absorption line for the cold-deposited and annealed films, is (0.3 ± 0.1) mm/sec. Summary data for the shift of the absorption line Δv_p and its broadening $\Delta\Gamma_{1/2}$ for all measured films as a function

are presented in Fig. 3 as a function of their thickness. After annealing, Δv_p and $\Delta\Gamma_{1/2}$ returned to zero. The thickness of the films was determined according to (3) from the absorption amplitude in the annealed film, whose absorption spectrum does not differ from the absorption spectrum in polycrystalline tin.

Referring to Fig. 2, we note that the absorption amplitude is practically the same for freshly deposited and annealed films, but the area of the curve, proportional to the observed half-width, is smaller for the annealed film and, consequently, the probability of absorption in the annealed film is smaller than in the freshly deposited one; in this case
$f'_{\text{cond}}/f'_{\text{anneal}} = (1.2 \pm 0.1)$.

Fig. 3. Summary data for the shift of absorption (I) and its broadening (II) in cold-deposited films. The dashed lines are obtained by processing the data by the method of least squares.

The observed deformation of the absorption spectrum in freshly deposited tin films, compared with the spectrum in annealed films, may be caused by several reasons.

First, for films less than 300 Å thick, the shift of the absorption peak may be associated with a new modification of tin that proves to be stable in thin layers

only at low temperatures. The possibility of the existence of a new modification in thin layers for a number of metals is reported in (4). The γ-modification of tin of orthorhombic symmetry is mentioned in (5). On the other hand, the Mössbauer spectrum in films more than 600 Å thick coincides with the spectrum in annealed films, while T_k remains the same as for thin layers with a deformed absorption spectrum. Apparently, the freshly deposited film is a mixture of two phases: the usual β-modification and a new modification stable only in thin layers. Depending on the relative fraction of each phase, either a deformed absorption spectrum for thin layers or the usual spectrum for thick films is observed. The critical temperature will then be determined by the modification with the higher T_k.*

Second, the broadening of the absorption curve for a freshly deposited film may be connected with a change in the electric-field gradient due to large stresses or packing defects in the film after its condensation onto a cold substrate. In addition, such stresses may lead to a change in the electron density at the nucleus and, consequently, to a change in the isomer shift. According to the data of (7), under all-round compression the Mössbauer spectrum in white tin is deformed, namely: the probability of absorption increases, and the absorption peak shifts linearly toward lower energies relative to the emission line of the source from Sn^*O_2, with a pressure shift derivative of $2.5 \cdot 10^{-6}$ mm/sec $\cdot$ atm.

* At one time, X-ray and electron-diffraction studies of freshly condensed tin films did not show the presence of lines of another structure (6). However, the X-ray studies were carried out on relatively thick films, and in electron-diffraction photographs local heating by the electron beam could cause a transition to the ordinary phase.

The stresses arising upon condensation in the film are opposite in sign to compressive stresses, and therefore the shift of the absorption line for films should be toward higher energies, which is indeed observed in our experiments. From the magnitude of the shift obtained, (0.20 ± 0.06) mm/sec, one can estimate the magnitude of the tensile stress, equivalent to a pressure of (80 ± 25) kbar, and the corresponding change in T_k; using the value $dT_k/dp = -4.5 \cdot 10^{-6}$ deg/atm for tin, we obtain $(3.6 \pm 1.5)^\circ$K. This is almost 4 times greater than the maximum value observed experimentally.

Third, the observed deformation of the absorption spectrum in chaotically oriented systems may be caused, according to ([8]), by anisotropy of the absorption probability in combination with a large quadrupole interaction in freshly deposited films. In our case, the calculated absorption curves, averaged over the direction of emission of γ-quanta relative to the symmetry axis c, can be brought into agreement with the experimental curves if the quadrupole-interaction parameter δ_a is taken to be (4.0 ± 0.5), and the anisotropy parameter σ_a to be (0.45 ± 0.10), corresponding to temperatures $\geq 300^\circ$K ([9]). If, however, one uses the data for absorption anisotropy at low temperatures ([2,9]), then the calculated curves differ little from the experimental curves in the polycrystal and, thus, cannot be made consistent with the results of measurements on freshly

deposited films.

Finally, the change in the absorption probability $\Delta f'/f'$, equal to (0.2 ± 0.1), makes it possible to estimate the possible change in the Debye parameter $\Delta\Theta/\Theta$, equal to (0.6 ± 0.3). The corresponding change in T_k, if one assumes the validity in this case of the BCS formula and does not take into account possible changes in the density of electronic states and in the electron-phonon interaction constant, is obtained as $(2 \pm 1)^\circ$K. This value is in full agreement with the observed change in T_k, if one takes into account the roughness of the estimate made.*

It remains to note that the least contradictory explanation of the deformation of the Mössbauer spectrum obtained experimentally is, apparently, a polymorphic transformation in the thin freshly condensed layer and the corresponding increase in T_k caused by a change in the Debye parameter. The influence of mechanical stresses, although also probable, does not make it possible to explain the constancy of T_k observed over a wide range of thicknesses, since in work ([11]) a dependence of the increase of T_k on the degree of deformation was shown. Moreover, large tensile stresses would seem to have to cause a decrease in the Debye parameter, whereas its increase is observed.

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* The assumption concerning the applicability of the usual BCS formula is confirmed by measurements of the isotope effect in freshly condensed tin films ([10]).

Note: Figure translations are in progress. See original paper for figures.