NMR double γ resonance in tantalum

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Absorption spectra were measured for Mössbauer radiation of Ta^{181*} (W) in tantalum under conditions when the nuclei are influenced by the source in a constant 3400-Oe magnetic field, by a variable magnetic field with a resonance frequency of 3 MHz for the excited state and intensities of 150, 300, and 360 Oe, by a field with a frequency of 4 MHz and an intensity of 300 Oe, and without a variable field. A theoretically predicted, sharp variation of the spectrum in 300- and 360-Oe fields at the resonance frequency, indicates that the NMR γ resonance is present.

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In Ta^{181} that has a Mössbauer level with a lifetime $\tau \approx 10^{-5}$ sec and having a large magnetic moment of the ground and isomeric states, a direct excitation of NMR double-γ resonance by an rf field is possible without an additional increase of the field at the nucleus. (For Fe^{57}, for example, an additional increase of $\sim 10^2 - 10^3$ is necessary.[1–3]) The absence of magnetostrictive oscillations in tantalum removes one of the fundamental difficulties of such experiments with magnetically ordered materials. To clearly observe the double resonance effect, it is necessary at a constant field intensity of $\sim(3-4)\times 10^3$ Oe to excite the Ta^{181} nuclei by a resonance variable field (frequency $\sim$MHz) with an intensity of several hundred Oersted in the source or in the absorber.

The first attempt to observe a double resonance in Ta^{181} at a constant field intensity $H_0 = 1800$ Oe and a variable field intensity (field amplitude) $H_{\sim} = 10$ Oe acting on the source was made by Kaindl and Salomon.[4]

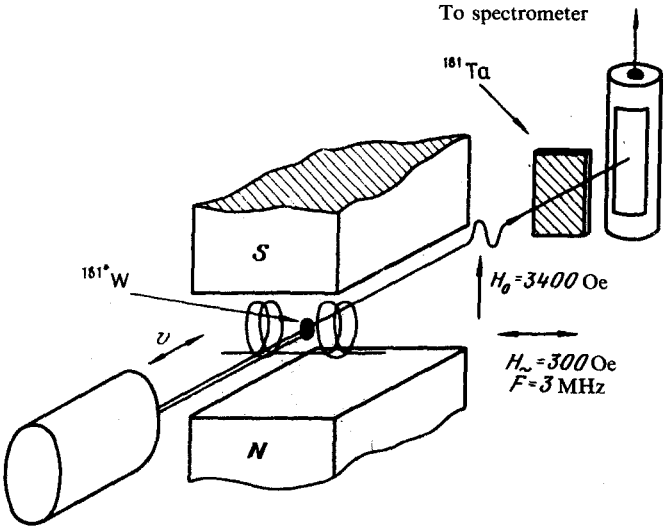

FIG. 1. Experimental setup.

In this paper we give the results of experiments in which at $H_0 = 3400$ Oe, the Mössbauer spectra of Ta^{181*} were analyzed by using a tantalum absorber in which resonance rf fields for the excited state (frequency $F = 3$ MHz) $H_\sim \approx 150$, $H_\sim \approx 300$, $H_\sim \approx 360$ Oe, and a field with $F = 4$ MHz and $H_\sim \approx 300$ Oe act on the nuclei of the source, and in which a double resonance in tantalum was observed.

The experimental setup is shown in Fig. 1. The Ta^{181*} source of Mössbauer radiation (W^{181*} in a tungsten matrix) was placed in constant and variable magnetic fields perpendicular to each other and to the direction of the analyzed radiation. The radiation transmitted through the absorber was detected by a proportional gas counter (Ar-Kr with some addition of CH_4).

Sources with 7–10 μCi activity (2×7 mm^2) were prepared by diffusion of metallic W^{181*} in a 10 to 30-μm-thick tungsten matrix. The W^{181*} was obtained by irradiation of a tantalum target in the internal deuteron beam of the FEI high-current cyclotron and, after separation from the original tantalum and elimination of the impurities in the ion-exchange columns (1×8 Dauex anionite), was reduced in a hydrogen flux in front of the metal. The 2-μm-thick tantalum foil, which was initially subjected to thermal processing, served as the absorber.

The constant magnetic field was generated by an electromagnet, and the rf was generated by a 12-mm-diam coil generator that contained eight turns of 2-mm-diam copper tubing through which water circulated. The power input to the coil reached 500 W, but the source, which was attached to the cooling rod of an electrodynamic vibrator made of BeO, did not heat up significantly.

The Mössbauer spectra were measured in a constant acceleration regime; the zero-line parabolic distortions inherent in this regime did not exceed 0.1–0.2%. The variable magnetic field intensity was measured with a test coil.

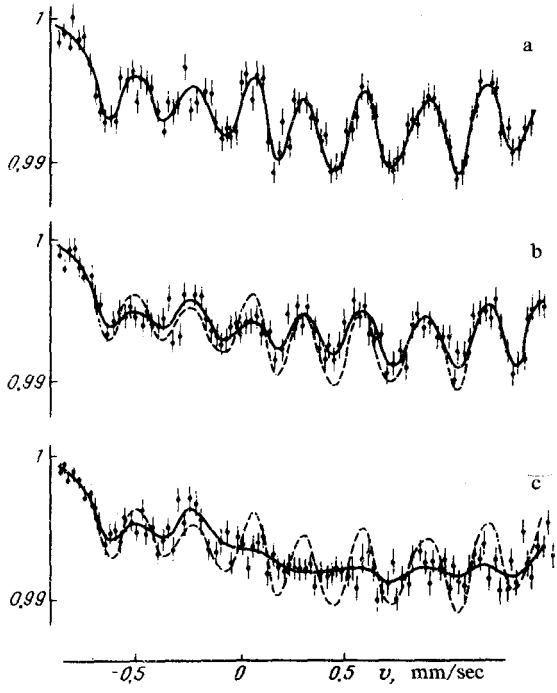

FIG. 2. Experimental Mössbauer absorption spectra of Ta^{181*} (W) radiation in tantalum at $H_0 = 3400$ Oe: a – $H_{\sim} = 0$; b – $F = 3$ MHz, $H_{\sim} \approx 150$ Oe; c – $F = 3$ MHz, $H_{\sim} \approx 300$ Oe. The dashed lines show the spectrum for $H_{\sim} = 0$.

Figure 2 shows experimental Mössbauer absorption spectra in tantalum (the total spectrum was not measured) at $H_0 = 3400$ Oe without a variable magnetic field (a) and in a variable field $H_{\sim} \approx 300$ Oe (c). The measurements were made in series alternately at $H_{\sim} \approx 150$ Oe, $H_{\sim} \approx 0$, and $H_{\sim} \approx 300$ Oe. The statistical accuracy of each spectral point is $\pm 0.1\%$.

The structure of the lines corresponding to the transitions from the Zeeman sublevels of the excited nuclear state-location and relative intensity - is shown in Fig. 4a. Not all the lines are resolved in the experimental spectrum (Fig. 2a), because of significant inhomogeneous broadening.

Under conditions of double resonance, when the frequency of the variable field is equal to the difference in the energies of the neighboring Zeeman sublevels of the excited state, at a sufficiently large intensity of the variable field each line (Fig. 4a) is split into $2I_E + 1$ components (I_E is the nuclear spin in the excited state).[5,6] The components of the lines are unresolved in the experimental spectra (Figs. 2b and 2c and Figs. 3b and 3d), but this splitting, which is proportional to $H_{\sim}$, produces a major change of the whole spectrum.

At $H_{\sim} \approx 150$ Oe (Fig. 2b) the amplitude of the lines in the middle region of the spectrum decreases significantly, and at $H_{\sim} \approx 300$ Oe (Fig. 2c) all the lines, except for the two on the extreme left-hand side, are almost completely smeared out. These variations are consistent with the computed spectra (Figs. 4b, 4c, and 4d). (Some discrepancy in the overall slope of the experimental and computed curves is attributable to the aforementioned small parabolic distortion of the zero line.)

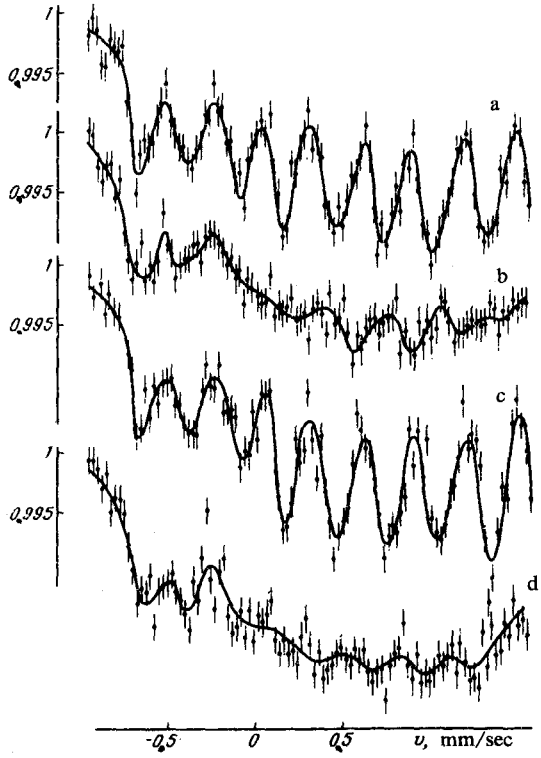

FIG. 3. Experimental Mössbauer absorption spectra for Ta^{181*} (W) radiation in tantalum at $H_0 = 3400$ Oe: $a - H_{\sim} = 0$; $b - F = 3$ MHz, $H_{\sim} \approx 360$ Oe; $c - F = 4$ MHz, $H_{\sim} \approx 300$ Oe; $d - F = 3$ MHz, $H_{\sim} \approx 300$ Oe.

Figure 3 shows the results of measurements using another source. In these experiments, additional discrimination of the pulses from the proportional counter was used over a period of time, which increased the resonance absorption effect to 18% for a line width $\Gamma = 0.14$ mm/sec. For a further increase of the resonance variable field intensity (Fig. 3b; computed curve Fig. 4e) there is an even sharper rearrangement of the spectrum, especially on the right-hand side: the peaks and valleys on the curve are shifted, so that a peak occurs at the location of the valley, and vice versa. This is due to the overlapping of the intense components of the split neighboring lines.

When a variable magnetic field acts on the nuclei of a source, in which the frequency of the field is shifted relative to the resonance $F = 4$ MHz, $H_{\sim} \approx 300$ Oe (Fig. 3c), the spectrum is almost the same as that at $H_{\sim} \approx 0$ (Fig. 3a). For comparison, we show in Fig. 3d the spectrum obtained with a variable field at the same intensity $H_{\sim} \approx 300$ Oe, but with the resonance frequency $F = 3$ MHz. (To increase the statistical accuracy, we used data for measurements with two sources.) From a comparison of Figs. 3a, 3c, and 3d it follows that the observed and the theoretically-predicted sharp variation of the spectrum in a variable magnetic field has a resonance nature.

In one of the experiments, we measured the Mössbauer absorption spectrum for the radiation of Ta^{181*} in tantalum, in which only a variable magnetic field ($F = 3$ MHz, $H_{\sim} \approx 300$ Oe) acted on the source nuclei, and compared it with the spectrum

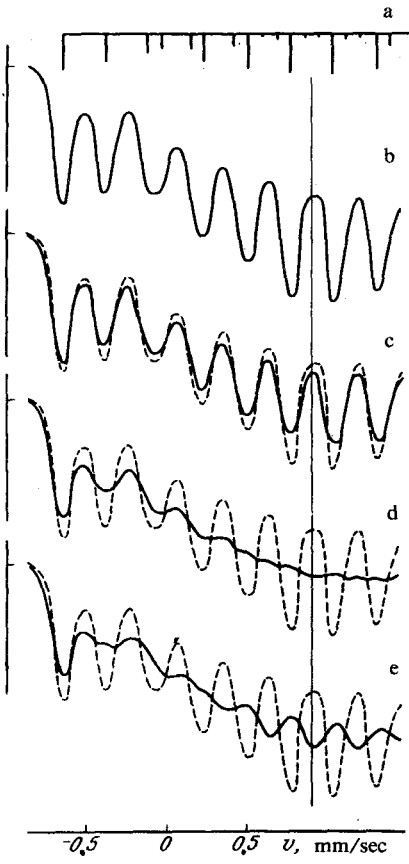

FIG. 4. (a) Structure of the radiation spectrum for ^{181}*Ta (W) (location and relative intensity of the lines) for $H_0 = 3400$ Oe, $H_\perp = 0$. The unsplit line location ($H_0 = 0$) is indicated by the vertical line. The computed Mössbauer absorption spectra for ^{181}*Ta (W) in tantalum for $H_0 = 3400$ Oe are: b—$H_\perp = 0$; c—$F = 3$ MHz, $H_\perp = 150$ Oe; d—$F = 3$ MHz, $H_\perp = 300$ Oe; e—$F = 3$ MHz, $H_\perp = 360$ Oe. The calculation was done for rigorous resonance conditions, in the thin-absorber approximation. The value $F = 3$ MHz may differ from that for the actual resonance frequency within a few percents, because of the imprecision of the magnetic moment of the excited state. The spectrum for $H_\perp = 0$ is represented by a dashed line.

obtained without the magnetic fields (single line). The absorption spectra are almost identical. This means that if the nuclear levels are not split by a constant magnetic field and, consequently, there are no resonance transitions between the Zeeman sublevels, the variable magnetic field does not affect the spectrum within the error of the measurements ($\pm 0.5\%$ at each point).

The obtained results indicate that the NMR double-γ resonance is present.

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[1]W. Meisel, Mössbauerspectrometrie, Proc. of the 4th Int'l. Conf. of Sco. Countries (1972).
[2]L. Pfeiffer, J. de Phys. **35**, C1-67 (1974).
[3]S. S. Yakimov, A. R. Mkrtchyan, V. N. Zarubin, D. V. Serbinov, and V. V.Sergeev, Pis'ma Zh. Eksp. Teor. Fiz. **26**, 16 (1977) [JETP Lett. **26**, 13 (1978)].

[4]G. Kaindl and D. Salomon, Bull. Am. Phys. Soc. **16**, 640 (1971); D. Salomon, Ph.D. Thesis (1972).
[5]M. N. Hack and M. Hamermesh, Nuovo Cim. **19**, 546 (1961).
[6]H. Gabriel, Phys. Rev. **184**, 359 (1969).