Frequency spectrum of a double nuclear magnetic γ resonance in Ta^{181}

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The frequency spectrum of the double nuclear magnetic γ resonance in the excited state of the source nuclei, Ta^{181*}, has been measured. The results are compared with calculations.

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The imposition of a strong rf magnetic field at an NMR frequency on Mössbauer nuclei splits the Zeeman nuclear sublevels and changes the structure of the Mössbauer spectrum.[1] The spectral lines split into $(2J_0 + 1)$ or $(2J_e + 1)$ components, where J_0 and J_e are the spins of the ground state and the excited state of the nucleus. The individual line components cannot be resolved experimentally, but the splitting changes the shape and amplitude of the observable lines which are of a resonant nature.

This effect—a double nuclear magnetic γ resonance (DMNGR)—has been observed experimentally in Ta^{181} nuclei.[2] A characteristic change in the shape of the Mössbauer spectrum was observed in Ref. 2 upon the application of a resonant rf field to nuclei of the radiation source, but there was no such change at a field frequency far from the NMR frequency.

In the present experiments we measured the DMNGR frequency spectrum, by which we mean the dependence of the intensity of an individual line in the Mössbauer spectrum on the frequency of the alternating magnetic field.

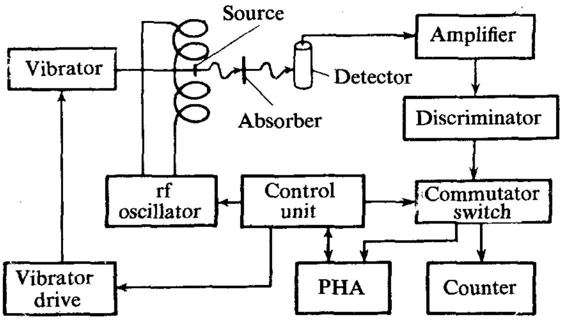

FIG. 1. Block diagram of the apparatus.

Figure 1 shows a block diagram of the DMNGR spectrometer. It consists of a Mössbauer spectrometer, an electromagnet, an rf oscillator with an exciting coil, and a control unit which maintains the appropriate operating conditions.

In frequency-sweep operation, the vibrator moves at a constant velocity, corresponding to the line of interest in the Mössbauer spectrum, while the frequency of the rf oscillator is tuned in accordance with the switching of the channels of the pulse-height analyzer (PHA). The capacitors of the oscillator circuit of the oscillator are switched by rf relays, which are controlled by the binary code of the address register of the pulse-height analyzer (32 channels are used). Synchronized adjustment of the plate voltage of the oscillator keeps the amplitude of the rf field from changing substantially as the frequency is tuned. The long-term frequency stability of the oscillator after heating is no worse than ±0.2%.

The experiments were carried out in an absorption geometry, with the rf magnetic field exciting transitions between the Zeeman sublevels of the excited state of the source nuclei, Ta^{181*} (W^{181}), in a static magnetic field 3330 (±0.5%) Oe. According to the calculations, the effect (the decrease in the absorption due to the splitting of the emission lines) should be greatest for the lines corresponding to the transitions $1/2 \rightarrow 1/2$ and $-1/2 \rightarrow -1/2$, which are thus the most convenient experimentally.

Figure 2 shows the DMNGR frequency spectra calculated (by the method of Ref. 1, without consideration of the effect of the ground state) for the $1/2 \rightarrow 1/2$ line for various amplitudes of the alternating magnetic field, $H_{\sim}$, along with the $H_{\sim}$ dependence of the magnitude of the effect and of the width of the spectral line. We see that the values of $H_{\sim}$ most convenient for observation lie in the interval of 0.2 — 0.3 of the experimental width of the Mössbauer line, $\Gamma_{\rm expt}$. The absorber used in the present experiments was of higher quality than that in Ref. 2 ($2\Gamma_{\rm expt} = 0.1$ mm/s); the measurements were taken at $H_{\sim} = 150 \pm 20$ Oe ($H/\Gamma_{\rm expt} = (0.3)$).[1]

Figure 3a shows the Mössbauer spectrum of Ta^{181} in a static magnetic field (the $1/2 \rightarrow 1/2$ and $-1/2 \rightarrow -1/2$ lines). The arrow shows the velocity at which the frequency spectrum was measured. The Mössbauer spectrum in the magnetic field and the frequency spectrum were measured in alternating series of measurements over the course of 2 months, so that the heights of the lines in these spectra could be compared quantitatively. The zero levels of the spectra were reconciled within ±0.15% through

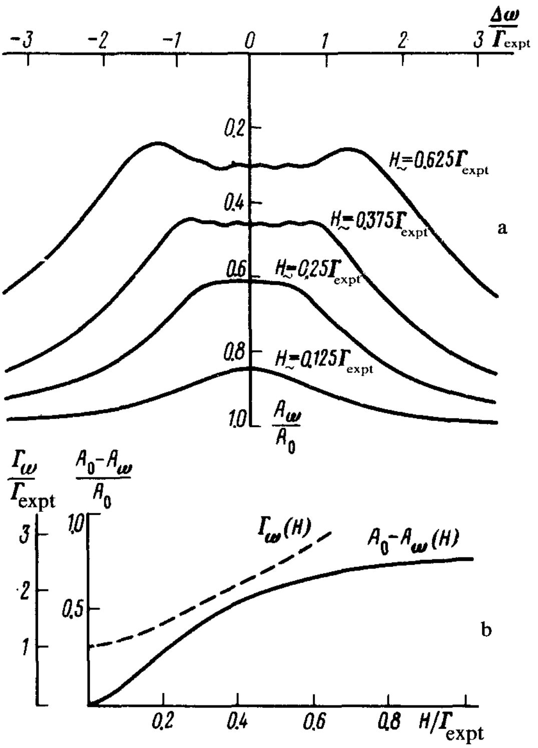

FIG. 2. a—Relative intensity of the line corresponding to the 1/2→1/2 transition vs the distance from the resonant frequency to the frequency of the alternating magnetic field, for various strengths of this field; b—relative width of a line in the frequency spectrum, $\Gamma/\Gamma_{\text{expt}}$, and magnitude of the DMNGR effect, $(A_0 - A_\omega)/A_0$ vs the strength of the alternating field.

a normalization based on a (negative) nonresonant velocity. The parameters of an individual line did not change over the time taken for the measurements ($2\Gamma_{\text{expt}} = 0.105 \pm 0.006$ mm/s, $\nu_0 = 0.773 \pm 0.006$ mm/s).

Figure 3b shows the DMNGR frequency spectrum measured over the range 2.2–4.6 MHz with a statistical error of ± 0.035% at each point in the spectrum. To check for possible parasitic effects (stray pickup from the high-power rf oscillator in the detection circuit, for example), we took measurements under frequency-sweep conditions. In one case, however, the differential discriminator was tuned to the x-ray lines of the Ta^{181*} source (8.15 and 9.34 keV), while in another it was tuned to the 6.25-keV Mössbauer line, but without an absorber. The statistical error was 0.025% at each point.

Gabriel[4] has derived a theory for DMNGR which takes into account the simultaneous effects of the rf field on the ground and excited states of the resonant nucleus. According to Ref. 4, each line in the Mössbauer spectrum generally splits into $(2J_e + 1)(2J_0 + 1)$ components in an rf field [as in Ref. 1, it is sufficient to consider the splitting into $(2J + 1)$ components in the immediate vicinity of the exact resonance]. Mitin[5] obtained some results which refine those of Ref. 4. According to Ref. 5,

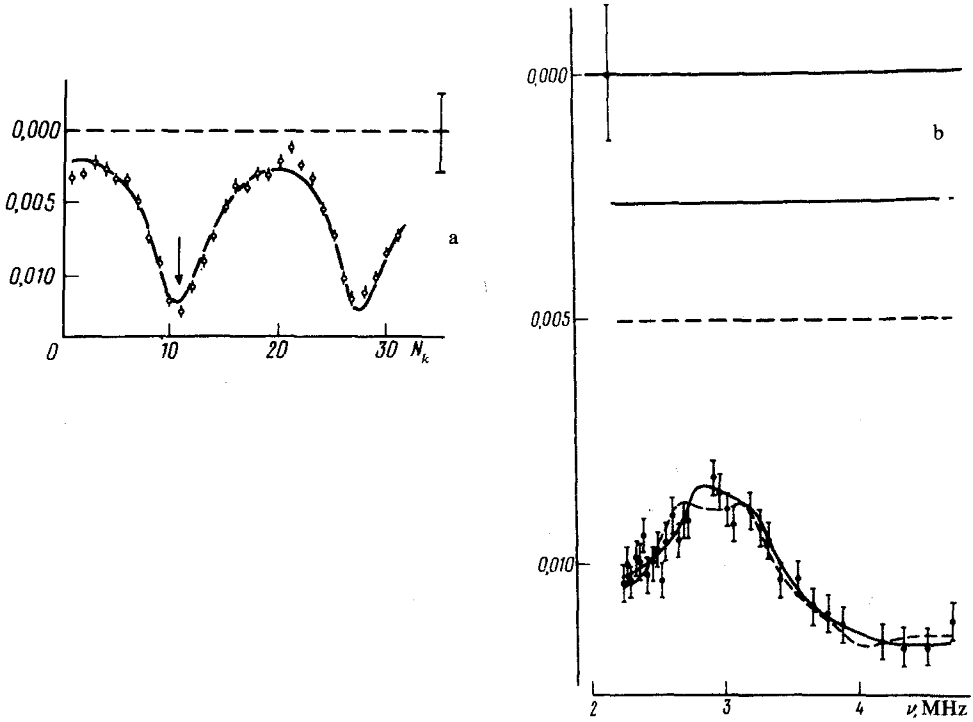

FIG. 3. Experimental Mössbauer spectra [$v = 0.0154\ (N_k + 32)$]. a—In a magnetic field ($H_0 = 3.33$ kOe, $n_k = 5.5\times 10^6$, $2\Gamma_{\rm expt} = 0.099 \pm 0.007$ mm/s, $v_0 = 0.780 \pm 0.003$ mm/s, $g_e/g_0 = 1.75 \pm 0.02$). The dashed line and the solid curve show the experimental and theoretical, respectively, positions of the nonresonant levels; b—experimental frequency spectrum for $H_0 = 3.33$ kOe, $v = 0.662$ mm/s, $2\Gamma_{\rm expt} = 0.1$ mm/s, and $n_k = 7.2\times 10^6$. Solid curve—calculated from Ref. 5 for $H = 163$ Oe and $g_e/g_0 = 1.76$; dashed curve—calculated from Ref. 4 for $H = 240$ Oe and $g_e/g_0 = 1.68$

the theory of Ref. 4 gives a generally correct description of the nature of the splitting, but incorporating the interference of the ground and excited states of the nucleus leads to changes in the amplitudes of the various components.

Figure 3b compares the experimental spectrum with the calculations of Refs. 4 and 5. In approximating the experimental points by the calculated lines, we adopted the following along with the scale of the curve as adjustable parameters: the zero level (the value found for it from the normalization of the spectrum within $\pm 0.5\%$ was included in the group of approximating experimental points, with the appropriate weight), the amplitude of the rf magnetic field (the accuracy in the experimental determination of this field by the various methods is no better than 10–15%), and the magnetic moment of the excited $\rm Ta^{181}$ isomer state (different, but not very different, values of g_e/g_0 were found in Refs. 6 and 7: between 1.75 and 1.80).

Comparison of the experimental spectrum with the calculations of Ref. 5 yields the ratio $g_e/g_0 = 1.76 \pm 0.07$, which agrees with the value $g_e/g_0 = 1.797 \pm 0.0007$ (the most accurate value), determined independently.[7] The comparison also yields $H_\sim = 160 \pm 90$ Oe, which is close to the nominal value (150 ± 20 Oe), and it yields zero levels for the theoretical and experimental spectra which are in better agreement. At the best approximation of the experimental spectrum with a calculation using[4] $H_\sim = 240 \pm 180$ Oe, the result, $g_e/g_0 = 1.68 \pm 0.12$, agrees with the value of 1.797 from Ref. 7 just barely within the experimental error. With $H_\sim = 160$ Oe (and with the other parameters optimized), there is a systematic discrepancy between the theoretical and experimental points on the high-frequency wing of the line. The experimental results thus seem to agree better with the calculations based on the theory of Ref. 5.

We are indebted to I. A. Semin for fabricating the Mössbauer source and to A. V. Mitin for a discussion of the DMNGR calculations.

[1] An attempt was made in Ref. 3 to observe the DMNGR frequency spectrum of Ta^{181} at the line 9/2→7/2 (for which the expected amplitude of the effect is four times smaller) at $H_\sim = 10$ Oe. Under these conditions, the total change in the line intensity is at least 60 times smaller than for the 1/2→1/2 transition at $H_\sim = 150$ Oe.

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[1] M. Hack and M. Hamermesh, Nuovo Cimento **19**, 546 (1961).
[2] V. K. Voĭtovetskiĭ, S. M. Cheremisin, and S. B. Sazonov, Pis'ma Zh. Eksp. Teor. Fiz. **30**, 711 (1979) [JETP Lett. **30**, 674 (1979)]; V. K. Voĭtovetskiĭ, S. M. Cheremisin, and S. B. Sazonov, Phys. Lett. **83A**, 81 (1981).
[3] G. Kaindl and D. Salomon, Bull. Am. Phys. Soc. **16**,. 640 (1971).
[4] H. Gabriel, Phys. Rev. **184**, 359 (1969).
[5] A. V. Mitin, Phys. Lett. **84A**, 278 (1981).
[6] G. Kaindl and D. Salomon, Phys. Lett. **32B**, 364 (1970).
[7] D. Salomon and P. West, Z. Phys. **A288**, 291 (1978).

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