Detection of Nuclear Magnetic Resonance on Oriented ^{193m}Pt*Fe* via the Anisotropy of Characteristic X Rays

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Nuclear magnetic resonance on oriented nuclei has been detected for the first time via the resonant change of the radiation pattern of characteristic L x rays. The magnetic hyperfine splitting $|g\mu_N B_{hf}/h|$ of $\frac{13}{2}^+$ ^{193m}Pt in Fe was determined to be 111.3(3) MHz. With $B_{hf} = -1261(25)$ kG, the g factor is deduced to be $(-)0.1158(23)$. This value differs strongly from the known g factors of $(\nu i_{13/2^+})^{-n}$ states in Hg, Pb, and Po, suggesting a significant change in the nuclear-deformation parameters of ^{193m}Pt.

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The technique of nuclear magnetic resonance on oriented nuclei detected via the anisotropy of γ radiation (NMR-ON)[1] has become a standard technique for the precise determination of the magnetic hyperfine splitting of radioactive nuclei in ferromagnetic host lattices, thus providing precise values for the magnetic moments of radioisotopes. For nuclear states decaying without significant γ emission the anisotropy of particles emitted can be used as the detector for NMR. In fact, conversion-electron-detected NMR has been reported for few isomeric states decaying only via highly converted transitions.[2,3] For the case of a pure electron-capture decay to the daughter ground state none of these methods will be applicable. On the other hand, the conversion process and the electron-capture decay are accompanied by the emission of *atomic* radiation. In this Letter we report on the first detection of NMR via the resonant change of the radiation pattern of characteristic x rays (X-NMR-ON).

In the decay modes of internal conversion (IC) and electron capture (EC) the polarization and alignment of the nuclei are transferred to the atomic system. The creation of polarized and/or aligned electronic vacancies causes a circular polarization and, if electronic levels with $j \geq \frac{3}{2}$ are involved, an anisotropy of the x rays emitted subsequently. An x-ray anisotropy can thus be expected for the L_3, M_3, M_4. . . radiation following an IC or EC process into the $2p_{3/2}$, $3p_{3/2}$, $3d_{3/2}$. . . atomic sublevels. Such an anisotropic emission of characteristic L_ι $(3s_{1/2} \rightarrow 2p_{3/2})$ and L_α $(3d_{3/2,5/2} \rightarrow 2p_{3/2})$ x rays has recently been observed for the first time in the decay of ^{191m}Ir*Fe* oriented at 15 mK.[4] Here we show that the L_ι and L_α anisotropies can be used as detectors for NMR on ^{193m}Pt*Fe*. A short description of preliminary results has been given elsewhere.[5]

The decay scheme of ^{193m}Pt is shown in Fig. 1(a). The 135.5- and 12.6-keV transitions are highly converted (γ-emission probability 0.1% and $< 1\%$, respectively). Suppose now that ^{193m}Pt is oriented completely. Then only the $m_I = +\frac{13}{2}$ substate will be populated, as indicated in Fig. 1(a). For the $M4$ decay to the $\frac{5}{2}^-$ level, conservation of angular momentum requires $\Delta I = 4$ and $|\Delta m_I| \leq 4$. Hence, for the com-

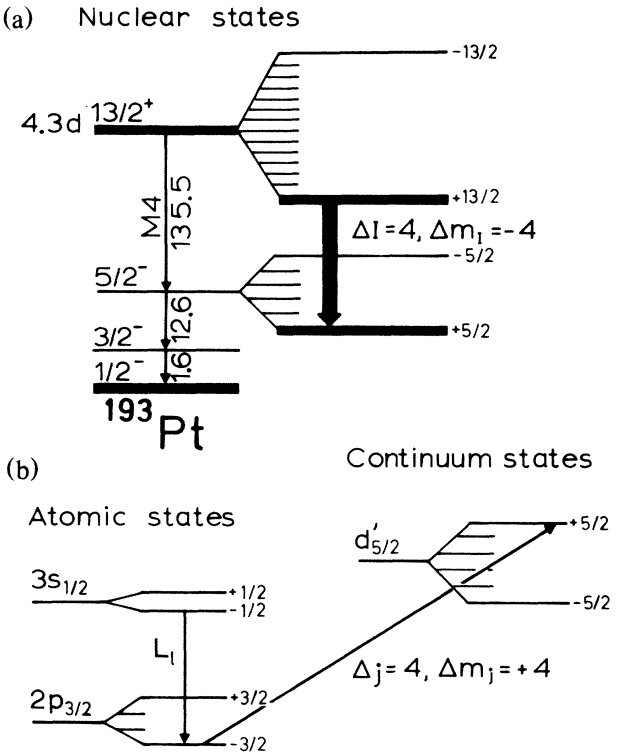

FIG. 1. (a) Decay scheme of ^{193m}Pt. Right-hand side: For completely oriented ^{193m}Pt only the $m_I = +\frac{13}{2}$ sublevel is populated. For the 135.5-keV $M4$ transition to the $\frac{5}{2}^-$ state $\Delta I = 4$ requires $|\Delta m_I| \leq 4$. Thus only transitions to the $m_I = +\frac{5}{2}$ substate are allowed, corresponding to $\Delta m_I = -4$. (b) With the constraint $\Delta j = 4$, $\Delta m_j = +4$ for the conversion electron transition only transitions from the $|2p_{3/2}, m_j = -\frac{3}{2}\rangle$ atomic sublevel to the $|d'_{5/2}, m_{j'} = +\frac{5}{2}\rangle$ continuum state are allowed in the 135.5-keV L_3 conversion of completely aligned ^{193m}Pt. Hence an aligned vacancy is created in the $2p_{3/2}$ shell and the following L_ι x rays are emitted anisotropically.

pletely aligned $\frac{13}{2}^+$ state, only $\Delta m_I = -4$ is allowed. If the decay energy is transferred to an electron with total angular momentum j and magnetic quantum number m_j the possible continuum states $|j'm_{j'}\rangle$ are confined by the condition $|j-4| \leq j' \leq j+4$ and $m_j' = m_j + 4$. In the decay of ^{193m}Pt 0.38 of a vacancy is created directly in the atomic $2p_{3/2}$ level by the 135-keV $M4$ conversion process. The allowed final continuum states for the conversion electron are $d'_{5/2}$, $g'_{7/2,9/2}$, and $i'_{11/2}$. (Transitions to $f'_{5/2,7/2}$ and $h'_{9/2,11/2}$ are forbidden because of parity conservation.) As the relative transition probabilities to $d'_{5/2}$, $g'_{7/2,9/2}$, and $i'_{11/2}$ are 1, 4×10^{-3}, and 3×10^{-4},[6] respectively, only the transitions to $d'_{5/2}$ have to be considered. Because of $\Delta m_j = +4$ only transitions from $|2p_{3/2}, m_j = -\frac{3}{2}\rangle$ to $|d'_{5/2}, m_{j'} = +\frac{5}{2}\rangle$ are allowed. Hence, an aligned $2p_{3/2}$ vacancy is created and the following L_3 x rays will be emitted anisotropically. The anisotropy is slightly reduced by the isotropic emission of L_3 x rays following the 135.5-keV K conversion and by the nearly isotropic emission following the 12.6-keV L_3 conversion. The 135.5-keV L_3 conversion and the following L_ι transition is illustrated in Fig. 1(b). The anisotropy of x rays is given by

$$W(\theta) = 1 + B_2 F_2 R_2 P_2(\cos\theta). \quad (1)$$

Here F_2 is the angular correlation coefficient for the observed atomic transition. The parameter R_2 describes the transfer of the nuclear alignment to the atomic vacancies created by the IC process; it depends on the spins and multipolarities of the involved nuclear transitions, on the partial matrix elements for the IC to all allowed final electron continuum states, and on preceding unobserved atomic transitions including Coster-Kronig transitions.[6,7] If the nuclear decay scheme is known, R_2 can be calculated with use of computer codes for partial internal conversion matrix elements and theoretical atomic transition probabilities.[8,9] The parameter B_2 describes the alignment of the initial nuclear state; it depends on $h\nu_M/k_B T$, where T is the temperature of the system and ν_M is the magnetic hyperfine splitting frequency,

$$\nu_M = |g\mu_N B_{\rm hf}/h| + |g|\mu_N \mathrm{sgn}(B_{\rm hf})(1+K)B_{\rm ext}/h. \quad (2)$$

Here g is the nuclear g factor, $B_{\rm hf}$ and $B_{\rm ext}$ are the magnetic hyperfine field and the external magnetic field, respectively, and K is a parameter taking into account Knight shift and diamagnetic shielding. The hyperfine splitting can be determined either via the temperature dependence of $W(\theta)$ or, as in the present case, much more precisley via the resonant change of $W(\theta)$.

The ^{193m}Pt*Fe* sample was prepared in the following way: ^{192}Pt (isotopic enrichment 52%) was melted with highly pure iron with a Pt concentration of 1 at.%. Foils with an area of 4×8 mm^2 and a thickness of 2 μm were produced by cold rolling and were neutron irradiated in the Julich reactor for 10 d in a neutron flux of 10^{14} $n/\mathrm{cm}^2\cdot\mathrm{s}$. After the irradiation two foils were annealed for 3 h at 650 °C in high vacuum ($\lesssim 10^{-6}$ Torr), soldered with In to the Ag cold finger of a demagnetization cryostat, and cooled to ~ 15 mK. The cold finger was tilted at 15° with respect to the direction of the external magnetic field, which is necessary to orient the ferromagnetic domains and to establish a unique direction of the hyperfine field. The angle of 15° was chosen as a compromise between maximal anisotropy at 0° and minimal self-absorption of the L x rays in the sample at 90°. The cryostat had been equipped with Be windows to achieve a high x-ray transmission at energies down to 8 keV.

For the most sensitive L x rays the following anisotropies were observed at $T = 17(3)$ mK: For L_ι ($3s_{1/2} \rightarrow 2p_{3/2}$; $E = 8.3$ keV),

$$W(15°) = 1.049(14);$$

for L_α ($3ds_{3/2,5/2} \rightarrow 2p_{3/2}$; $E = 9.4$ keV),

$$W(15°) = 1.0073(12).$$

In the X-NMR-ON experiment the rf field was applied with a two-turn coil perpendicular to $B_{\rm ext}$. The resonance was searched for between 90 and 120 MHz. Figure 2 shows the X-NMR-ON spectra of the L_ι and L_α transitions accumulated for seven demagnetization runs with a total measurement time of 7 d. The observed resonance amplitudes represent a 50% destruction of the x-ray anisotropies.

When we take into account the extrapolation to zero external polarizing field, the magnetic hyperfine splitting frequency of ^{193m}Pt*Fe* is found to be

$$|g\mu_N B_{\rm hf}/h| = 111.3(3)\ \mathrm{MHz}.$$

With the known hyperfine field of $-1261(25)$ kG [data of Kontani and Itoh[10] recalculated with $\mu(^{195}\mathrm{Pt}) = 0.609\,49(6)\mu_N$ of Lederer and Shirley[11]] the g factor and the magnetic moment of $\frac{13}{2}^+$ ^{193m}Pt are deduced to be

$$g = (-)0.1158(23), \quad \mu = (-)0.753(15)\mu_N,$$

where the negative sign has been taken from the known systematics. By application of the empirical rule of Moskowitz and Lombardi,[12] which has meanwhile been founded theoretically by Fujita and Arima,[13] and hyperfine anomaly between ^{195}Pt and ^{193m}Pt is estimated to be 0.3%, which is small in comparison to the uncertainty introduced by the error of the hyperfine field and can thus be neglected.

The g factor of ^{193m}Pt can be compared with the known g factors of the isomeric $(\nu i_{13/2})^{-n}$ states in ^{195}Pt,[14] Hg,[15] Pb,[16] and Po,[17] which are illustrated in

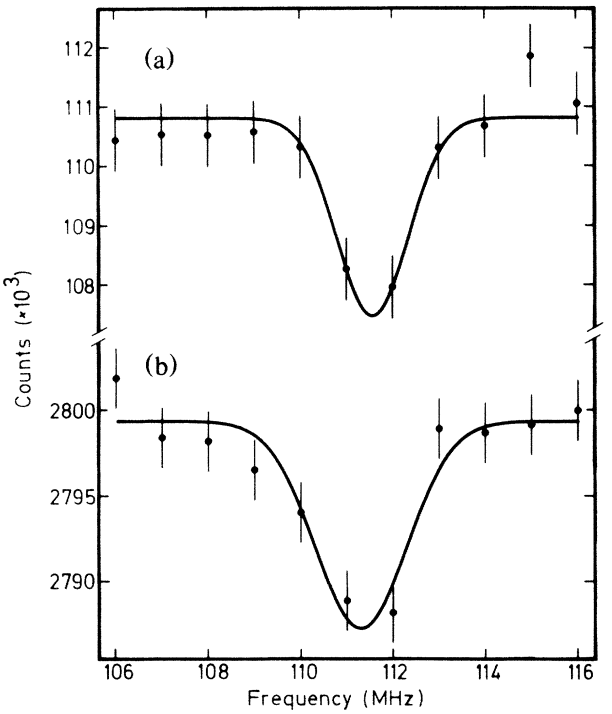

FIG. 2. X-NMR-ON spectra of (a) the L_ι transition and (b) the L_α transition of ^{193m}Pt*Fe*, measured in an external magnetic field of 1 kG during the warmup of the cryostat from 15 to 20 mK. About 50% of the x-ray anisotropy is destroyed. Total accumulation time seven days.

Fig. 3. While the g factors in Hg, Pb, and Po show only a smooth variation in a wide range of the neutron number, there is a drastic change for both Pt isotopes. The magnetic moments of the (nearly) spherical Hg, Pb, and Po isomers are well described by the modified Schmidt formula[18]

$$\frac{\mu^{\text{eff}}}{\mu_N} = I\left\{g_l^{\text{free}} + \delta g_l \pm \frac{(g_s^{\text{free}} + \delta g_s) - (g_l^{\text{free}} + \delta g_l)}{2I+1}\right\} + \text{tensor term}, \quad (3)$$

where δg_l is a state-independent correction to the free-nucleon orbital g factor g_l^{free}, and δg_s is a state-dependent correction to the free-nucleon spin g factor g_s^{free} mainly due to core polarization, the contribution of the tensor term being small. The data of Fig. 3 indicate only a small dependence of the core polarization on the neutron number and on the proton number. The large deviation of the Pt g factors cannot be due to core polarization as pointed out by Bacon *et al.* for the explanation of the magnetic moment of ^{195m}Pt.[14]

On the other hand it is known that the heavier Pt nuclei with $A > 189$ are moderately deformed with oblate shapes. In the framework of strong coupling of the single particle to a well deformed core the magnetic moment is given by[19]

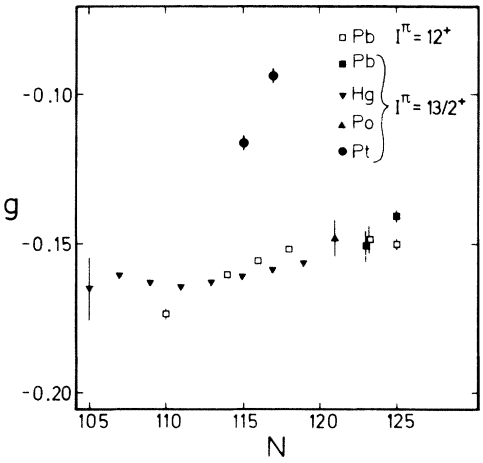

FIG. 3. Experimental g factors of the $(\nu i_{13/2})^{-1}$ isomers in ^{193}Pt (this work), ^{195}Pt (Ref. 14), Hg (Ref. 15), Pb (Ref. 16), Po (Ref. 17), and the $(\nu i_{13/2})^{-2}$ isomers in Pb (Ref. 16) as a function of the neutron number N.

$$\mu/\mu_N = g_R I + [K^2/(I+1)](g_K - g_R), \quad (4)$$

where g_K and g_R are the effective single-particle g factor and the collective g factor, respectively. In this way, with $g_R = +0.35(5)$ and $g_K K = -1.0(1)$, $\mu = -0.56(13)\mu_N$ is calculated for a $I^\pi K = \frac{13}{2}^+\frac{13}{2}$ state, nearly independent of the deformation. The good agreement with $\mu = -0.605(15)\mu_N$ for ^{195m}Pt [Ref. 14, recalculated with $B_{\text{hf}} = -1261(25)$ kG] may be somewhat accidental. It indicates, however, that the different magnetic moments of ^{193m}Pt and ^{195m}Pt may be understood as being caused by static or dynamic deformations, which seem to vary strongly with the neutron number. The effects should be large enough to stimulate theoretical work for the understanding of this behavior.

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