Evidence for a Strong Impurity-Nucleus Nuclear-Magnon Interaction of ^{89m}Y in hcp Cobalt

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We report the first observation of a nuclear-magnetism contribution to the magnetic hyperfine interaction of impurity atoms in a ferromagnetic host lattice. For $^{89m}YCo^{(\mathrm{hcp})}$ a significant temperature dependence of the magnetic hyperfine interaction was observed for $10 \leq T \leq 25$ mK. It is attributed to a strong coupling of the ^{89m}Y nuclei to collective nuclear-magnon excitation modes of the ^{59}Co nuclear spin system. [S0031-9007(97)04064-7]

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It is well known that a magnetic hyperfine field B_{HF} exists at the nuclear site of atoms dissolved as dilute impurities in a ferromagnetic host lattice such as Fe, Co, Ni, Gd [1]. The magnetic hyperfine interaction frequency is given by

$$\nu_0 = \nu_M + |g| \mu_N/h(1 + K)B_{\mathrm{ext}} \operatorname{sgn}(B_{\mathrm{HF}}), \quad (1)$$

$$\nu_M = |g\mu_N B_{\mathrm{HF}}/h|, \quad (2)$$

where g is the nuclear g factor, B_{ext} is an external magnetic field, $\operatorname{sgn}(B_{\mathrm{HF}})$ is the sign of B_{HF} with respect to B_{ext}, and K is a parameter including Knight shift and diamagnetic shielding.

The hyperfine field is proportional to the electronic magnetization $\vec{M}$, $\vec{B}_{\mathrm{HF}} \propto \vec{M}$. M is temperature dependent, $M(T) = M(T = 0)(1 - AT^{3/2})$, where A is a constant which is of the order $10^{-6}\,\mathrm{K}^{-3/2}$ for Fe, Co, and Ni. For $T < 100$ mK the hyperfine fields differ from the saturation values by $\lesssim 10^{-6}$. It is thus evident that, for this temperature region, no temperature dependence of the magnetic hyperfine splitting has been observed until now.

The question of whether *nuclear-magnetization* effects yield a contribution to the magnetic hyperfine interaction of dilute impurities in ferromagnets has, to our knowledge, not been addressed until now. In this context the question arises of whether a coupling of impurity nuclei to nuclear-magnon modes exists. In a recent experiment on ^{191}Pt in fcc Co it was found that the relaxation rate increased by 2 orders of magnitude, which was attributed to the existence of a nuclear-magnon-induced relaxation mechanism [2]. The resonance frequency of $^{191}PtCo^{(\mathrm{fcc})}$, however, was not affected. Nevertheless, a strong impurity-nucleus nuclear-magnon interaction, if existent, could influence the resonance frequency of the impurity nuclei strongly. As the basis for such an effect, if existent, a large nuclear magnetization m of the host lattice is essential. Thus Fe and Ni can be left out of consideration. Cobalt, however, is monoisotopic, and the nuclear magnetic moment of ^{59}Co is relatively large, $\mu = 4.6\mu_N$. For hcp Co the ratio of the nuclear saturation magnetization m_0 to the electronic saturation magnetization M_0 is $m_0/M_0 = 1.5 \times 10^{-3}$. The nuclear magnetization of Co becomes relevant if the thermal energy is of the same order of magnitude as the $^{59}CoCo$ hyperfine splitting, i.e., for temperatures $T < 100$ mK. Therefore the experiments described here were performed with hcp Co as host lattice, at temperatures well below 100 mK.

In magnetically ordered solids such as ferromagnetic Co an indirect coupling between like nuclear spins I exists via the hyperfine interaction $A\vec{I} \cdot \vec{s}$ and the virtual excitation and absorption of electronic spin waves—the Suhl-Nakamura (SN) interaction [3,4]. For $T < 100$ mK this coupling leads to a drastic change in the excitation behavior of the nuclear-spin system. Instead of single-spin flips the elementary excitations are collective, i.e., nuclear magnons. This is connected with a considerable reduction of the magnetic hyperfine interaction frequency, the so-called "frequency pulling" [5].

The "pulled" frequency ν_p is to first-order approximation given by [5,6].

$$\nu_p = \nu_0 - \delta\nu_p, \quad (3)$$

$$\delta\nu_p = \nu_M \frac{\eta_x + \eta_y}{2} \frac{m_0}{M_0} \frac{\langle I_z \rangle}{I}. \quad (4)$$

Here ν_0 is the "intrinsic" (without coupling by the SN interaction) magnetic hyperfine interaction frequency as given by Eq. (1), $\delta\nu_p$ is the frequency pulling, $\eta_{x,y}$ is the enhancement factor in x, y direction—it takes into account the enhancement of an external magnetic field at the nuclear site by the hyperfine interaction; the x-y plane is perpendicular to the orientation axis—m_0 and M_0 are the saturation values of the nuclear and electronic magnetization, respectively, and $\langle I_z \rangle / I$ is the fraction of nuclear polarization of the ^{59}Co spin system.

The enhancement factor $\eta_{x,y}$ depends on the magnitude and the direction of the external magnetic field and on the sample geometry. For the special case B_{ext} parallel to the c axis of Co, $\eta_{x,y}$ is given by

$$\eta_{x,y} = \frac{B_{\mathrm{HF}}}{B_{\mathrm{ext}} + B_A^{(\|)} + (N_{x,y} - N_z)\mu_0 M_0}, \quad (5)$$

where $B_{\mathrm{A}}^{(\|)} = 10.6$ kG is an anisotropy field, and $N_{x,y,z}$ are the demagnetization factors in the x, y, z direction. The saturation magnetization of Co is $\mu_0 M_0 = 18.1$ kG. The frequency pulling depends on the external magnetic field (via $\eta_{x,y}$) and on the temperature (via $\langle I_z \rangle / I$). Recently,

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this frequency pulling has been observed for hcp Co in the temperature region 10 . . . 30 mK [6]: Experimental values for the pulled and unpulled frequency of ^{59}Co in hcp Co are $\nu_p = 215 \ldots 218$ MHz ($T = 10 \ldots 30$ mK) and $\nu_0 = 219.9$ MHz ($B_{\rm ext} = 0$). The pulled and unpulled frequencies ν_p and ν_0 represent the excitation energies of the $k = 0$ and $k \gg 1/b$ nuclear magnons (b is the range of the SN interaction). Thus, the nuclear-magnon spectrum lies between ~215–220 MHz.

The question of whether the collective nuclear magnetism of ^{59}Co has an influence on the magnetic hyperfine interaction of impurity nuclei cannot be answered *a priori*. It could be speculated, however, that the effect, if existent, should be large if the hyperfine interaction of the impurity nuclei is within the energy band given by the nuclear-magnon dispersion relation. Therefore we studied the system $^{89m}\mathrm{Y}Co^{(\mathrm{hcp})}$, which meets this criterion.

The hyperfine interaction of $^{89m}\mathrm{Y}Co^{(\mathrm{hcp})}$ can be predicted taking into account the results from NMR-ON measurements on $^{91m}\mathrm{Y}Co^{(\mathrm{hcp})}$ [7] and $^{89m,91m}\mathrm{Y}Fe$ [8]: Taking $\nu_M^{(\parallel)}(^{91m}\mathrm{Y}Co^{(\mathrm{hcp})}) = 208.89(3)$ MHz [8], $\nu_M^{(\perp)}(^{91m}\mathrm{Y}Co^{(\mathrm{hcp})}) = 200.41(57)$ MHz [7], and $g(^{89m}\mathrm{Y})/g(^{91m}\mathrm{Y}) = 1.0441(2)$ [8], $\nu_M^{(\parallel)}(^{89m}\mathrm{Y}Co^{(\mathrm{hcp})}) = 218.10(5)$ MHz and $\nu_M^{(\perp)}(^{89m}\mathrm{Y}Co^{(\mathrm{hcp})}) = 209.25(60)$ MHz is obtained. Here $\parallel$ and $\perp$ denote parallel and perpendicular magnetization with respect to the crystal c axis.

In general, the magnitude of the hyperfine field in hcp Co depends on the angle θ between the electronic magnetization $\vec{M}$ and the crystallographic c axis:

$$B_{\rm HF}(\theta) = B_{\rm HF}^{\parallel}\cos^2\theta + B_{\rm HF}^{\perp}\sin^2\theta = B_{\rm HF}^{(\rm iso)} + B_{\rm HF}^{(\rm anis)}P_2(\cos\theta)\,. \quad (6)$$

The anisotropy of the hyperfine field is element specific: For Co and Y in hcp Co this anisotropy has opposite sign: $\mathrm{Co}Co^{(\mathrm{hcp})}$: $B_{\rm HF}^{(\rm anis)} = +5.4(1)$ kG [9]; $\mathrm{Y}Co^{(\mathrm{hcp})}$: $B_{\rm HF}^{(\rm anis)} = -5.5(4)$ kG [7]. This fact allows one to adjust the difference between the intrinsic magnetic hyperfine interaction of ^{89m}Y and the $k = 0$ nuclear-magnon frequency. For $\vec{M} \parallel c$ this difference can be adjusted to vanish; for $\vec{M} \perp c$ and $B_{\rm ext} = 20$ kG, this difference exceeds 15 MHz; for the case that the c axis is tilted 40° with respect to the direction of $B_{\rm ext}$, the frequency difference can be adjusted by the external magnetic field between -0.1 MHz (for $B_{\rm ext} = 2$ kG) and $+3.2$ MHz (for $B_{\rm ext} = 13$ kG).

Samples of $^{89m}\mathrm{Y}Co^{(\mathrm{hcp})}$ were prepared with the recoil-implantation technique: Targets consisting of a thin disk-shaped Co single crystal (thickness ~6 μm; c axis oriented parallel to the plane of the disk) and a 1.5 μm thick ^{90}Zr foil in front of it were irradiated with 55 MeV α particles at the cyclotron in Karlsruhe. In this way ^{89}Zr ($T_{1/2}$=78.4 h) is produced via the nuclear reaction $^{90}\mathrm{Zr}(\alpha, 5n)^{89}\mathrm{Mo} \rightarrow {}^{89}\mathrm{Nb} \rightarrow {}^{89}\mathrm{Zr}$ and a sufficient amount of $A = 89$ atoms is recoil implanted into Co. In the decay of ^{89}Zr the isomeric state $^{89m}\mathrm{Y}(T_{1/2} = 16.1$ s) is populated. As a by-product, the samples contained activities of ^{90}Nb, ^{56}Co, and ^{58}Co. After the irradiations, the samples were soldered to the coldfinger of a ^{3}He $-$ ^{4}He-dilution refrigerator and cooled down to a temperature of ~10 mK. A superconducting magnet supplied an external magnetic field $B_{\rm ext} = 0 \ldots 20$ kG. For three different types of experiments, the single-crystal c axis was adjusted parallel to the direction of $B_{\rm ext}$ (denoted as "0° geometry"), perpendicular to $B_{\rm ext}$ ("90° geometry"), and with an angle $\alpha = 40°$ between c and $B_{\rm ext}$ ("40° geometry"). The γ rays were detected with four Ge detectors placed at 0°, 90°, 180°, and 270° with respect to the direction of $B_{\rm ext}$. The temperature of the coldfinger was measured via the γ anisotropy of a $^{60}\mathrm{Co}Co^{(\mathrm{hcp})}$ nuclear thermometer. In addition, the temperature of the sample was controlled via the γ anisotropies of the contaminants ^{90}Nb, ^{56}Co, and ^{58}Co. The radio frequency (rf) for the NMR-ON measurements, supplied by a rf synthesizer, was applied either frequency modulated with a modulation band width 0.1 . . . 1 MHz ("FM on") or without FM ("FM off"), and γ-ray spectra were recorded as a function of the rf frequency. Typically, FM-on and FM-off spectra were measured consecutively.

First, we measured in 90° geometry ($c \perp B_{\rm ext}$). For this geometry, full magnetization is obtained for $B_{\rm ext} \geq B_{\rm A}^{(\perp)} = 13.4$ kG [10]. The upper part of Fig. 1 shows spectra for $B_{\rm ext} = 20$ kG, measured with FM (left) and without FM (right). The spectra show the regular

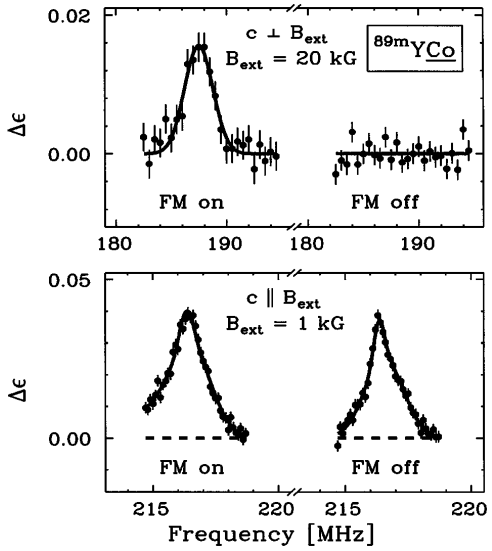

FIG. 1. NMR-ON on $^{89m}\mathrm{Y}\mathrm{Co}^{(\mathrm{hcp})}$: Change of the γ anisotropy $\Delta\epsilon$ of the 909 keV transition of ^{89m}Y vs frequency of the applied rf field. Here $\epsilon = W(0°)/W(90°)$, where $W(\vartheta)$ is the γ ray angular distribution. Top: 90° geometry ($B_{\rm ext} \perp c$); $B_{\rm ext} = 20$ kG. Here the "normal" behavior of NMR-ON resonances is observed. The resonance only can be observed with frequency-modulated rf (left), whereas there is no resonance signal for unmodulated rf (right). This is due to the inhomogenous broadening of the hyperfine interaction frequency. Bottom: 0° geometry ($B_{\rm ext} \parallel c$); $B_{\rm ext} = 1$ kG. Here a considerably strong resonance signal is obtained for unmodulated rf (right). This indicates the existence of an effect by which a large homogenous broadening is introduced. The frequency modulation band width was $\Delta_{\rm FM} = \pm 1$ MHz (90° geometry) and ± 0.25 MHz (0° geometry).

behavior of NMR-ON resonances in ferromagnets: Because of the low ^{89m}Y concentration of $\lesssim 10^{-10}$ the homogeneous broadening of the resonance ($\Gamma_{\text{hom}} \lesssim 100$ Hz) is much smaller than the inhomogeneous broadening ($\Gamma_{\text{inh}} \sim 2$ MHz). Thus, by applying the rf at a fixed frequency only a negligibly small fraction of ^{89m}Y nuclei is resonated. A considerably large resonance effect is obtained only if the rf is applied frequency modulated, with a FM band width which is comparable to the inhomogeneous broadening. The center frequency of the FM-on resonance is 187.56(12) MHz. It is in good agreement with 188.13(65) MHz, which is expected taking the data on ^{91m}Y in hcp Co and the known ratio of g factors [7,8].

Next, measurements were performed for 0° geometry ($c \parallel B_{\text{ext}}$), for which the hyperfine interaction frequency of ^{89m}Y is close to the nuclear-magnon excitation frequencies of ^{59}Co. Here, the resonance behavior of ^{89m}Y is completely different from all NMR-ON spectra observed so far. The FM-on and FM-off spectra for $B_{\text{ext}} = 1$ kG (lower part of Fig. 1) show that the resonance can be excited by unmodulated rf as well as by modulated rf. A simultaneous *least-squares* analysis of the FM-on and FM-off spectra, in which the FM modulation band width is unfolded for the FM-on spectra, yields essentially the same frequency distribution for FM on and FM off. It should be noted that this resonance behavior was exclusively found in the γ anisotropy of the 909 keV transition of ^{89m}Y and not in the γ anisotropies of ^{90}Nb, ^{56}Co, and ^{58}Co which served as internal thermometers. The frequency independence of the γ anisotropies of ^{90}Nb, ^{56}Co, and ^{58}Co, and of the ^{60}Co*Co* thermometer prove that the rf power level was chosen low enough so that the conditions for a calorimetric detection of the ^{59}Co resonance were not fulfilled. Thus it is guaranteed that the observed resonance must be attributed to NMR on ^{89m}Y. In addition, a homogeneous broadening of the ^{89m}Y*Co* hyperfine interaction must have been introduced, which we interpret as being due to the coupling of ^{89m}Y to the ^{59}Co nuclear-magnon modes.

Another extraordinary property of the ^{89m}Y resonance in 0° geometry can be seen in Fig. 2, which shows FM-off spectra measured for $T = 13$, 18, and 24 mK: There is a completely unexpected temperature dependence in the milli-Kelvin region. For all dilute impurities in ferromagnets investigated until now the temperature dependence of the magnetic hyperfine interaction originates from the reduction of the electronic magnetization with increasing temperature. For hcp Co and $T < 100$ mK the corresponding relative increase of B_{HF} with decreasing temperature should be well below 10^{-6}. In contrast, the hyperfine interaction frequency of ^{89m}Y is reduced between 24 and 13 mK by about 4×10^{-3}.

The ^{89m}Y resonance structure can be well described by a narrow ($\Gamma \sim 0.5$ MHz) and a broad ($\Gamma \sim 2$ MHz) resonance (full curves in Fig. 2). The resonance centers are nearly coinciding; the resonance maximum is given by the narrow resonance. For the existence of this particular resonance structure we cannot give a satisfactory explanation at present.

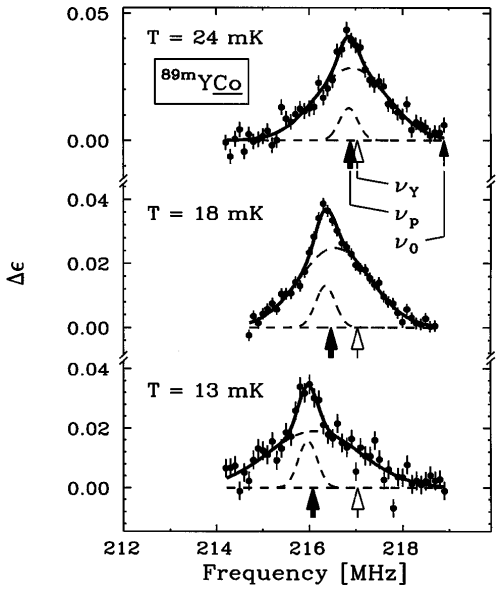

FIG. 2. "FM-off" spectra of ^{89m}Y$\text{Co}^{(\text{hcp})}$ measured for different temperatures. The intrinsic resonance frequency of ^{89m}Y (ν_{Y}) and the unpulled (ν_{o}) and pulled (ν_p) frequencies of ^{59}Co are marked with arrows. The resonance structure can be well described by the superposition of a narrow and a broad resonance with maximum at $\overline{\nu}$.

In Fig. 3 the frequency of the resonance maximum $\overline{\nu}$ is shown vs T (upper scale) and vs the corresponding fractional orientation $\langle I_z \rangle / I$ of ^{59}Co (lower scale). The solid and broken lines represent the pulled frequency ν_p of ^{59}Co and the intrinsic hyperfine interaction frequency ν_{Y} of ^{89m}Y, respectively. It is obvious that the magnetic hyperfine interaction frequency of ^{89m}Y is close to the ^{59}Co $k = 0$ nuclear-magnon frequency. This can be interpreted that the precession of ^{89m}Y is "synchronized" by the transverse magnetic field of the collective ^{59}Co mode.

Next, the question was addressed of how the resonance behavior of ^{89m}Y depends on the frequency difference between the intrinsic magnetic hyperfine interaction of ^{89m}Y and the ^{59}Co $k = 0$ nuclear-magnon frequency. This difference can be adjusted by three experimental parameters, T, B_{ext}, and the angle α of the single crystal c axis with respect to B_{ext}. This is due to the fact that the anisotropies of the hyperfine field [Eq. (6)] of Co and Y in hcp Co have opposite signs. We chose $\alpha = 40°$, as a compromise between a not too large reduction of the γ anisotropy in the direction of the 0° and 180° detectors (small α) and a large dynamical range for θ (large α). The results for the ^{89m}Y resonance maxima $\overline{\nu}$ are shown in Fig. 4. The dashed and solid lines represent the intrinsic resonance frequency ν_{Y} of ^{89m}Y and the pulled frequency ν_p of ^{59}Co, respectively. Obviously, the ^{89m}Y resonance frequency $\overline{\nu}$ lies between ν_{Y} and ν_p, relatively near to ν_p. The data suggest $\overline{\nu} - \nu_p \propto \nu_{\text{Y}} - \nu_p$. With increasing $\nu_{\text{Y}} - \nu_p$ the amplitude

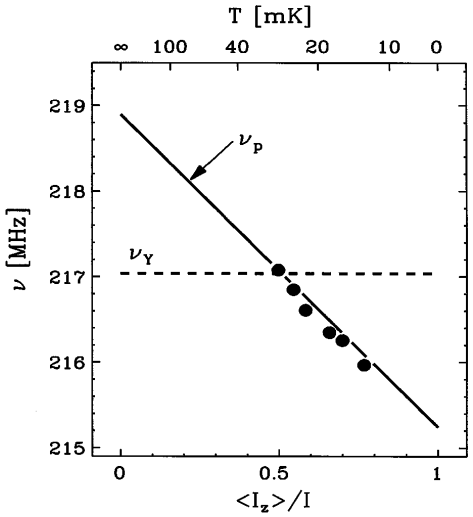

FIG. 3. Dependence of the "FM-off"-resonance maximum of ^{89m}Y ($\overline{\nu}$) on the fractional orientation of the ^{59}Co nuclear spin system (bottom scale) and the respective temperature (top scale). It follows well the frequency of the pulled ^{59}Co resonance (full curve).

of the resonance decreases; for $\theta > 20°$ ($|\nu_Y - \nu_p| > 2.8$ MHz) the resonance could not be observed any more.

In summary, we observed a rather unexpected behavior of the NMR-ON resonance behavior of ^{89m}Y*Co*. The resonance is homogeneously broadened and shifted in frequency towards the frequency of the collective Co excitation if the difference of frequencies does not exceed a critical value $\Delta_c \sim 3$ MHz. This critical frequency difference is larger than the Suhl-Nakamura (SN) width and the frequency pulling: According to Ref. [11] the SN width is estimated to be ~40 kHz. The frequency pulling at the "critical" point is $\delta\nu_p \sim 1.5$ MHz, i.e., only about half of Δ_c.

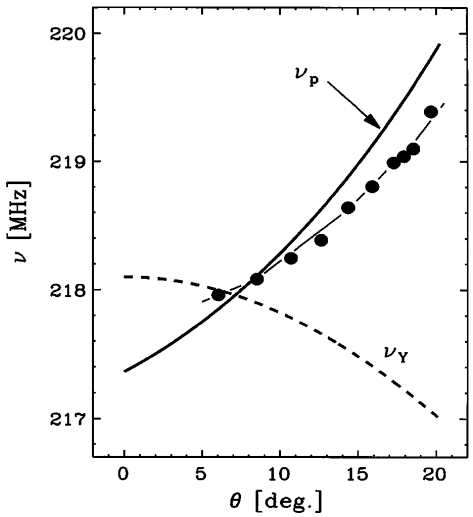

FIG. 4. "FM-off"-resonance maximum $\overline{\nu}$ of ^{89m}Y as function of θ (angle between the electronic magnetization and the single-crystal c axis) measured for $T = 20$ mK in 40° geometry. The angle θ was adjusted via the external magnetic field $B_{\text{ext}} = 2$ kG ($\theta = 6°$)...10 kG ($\theta = 20°$). For clarity of presentation the resonance shift caused by B_{ext} was subtracted. The theoretical resonance frequencies of ^{89m}Y (ν_Y) and the pulled ^{59}Co resonance frequency for $T = 20$ mK (ν_p) are represented by the dashed and the full line, respectively. The slender broken line indicates the experimental trend. The opposite trend of ν_Y and ν_p is due to the anisotropy of the hyperfine field, which has an opposite sign for Y and Co.

The following two different explanations can be offered: (i) The ^{59}Co nuclear magnetism produces a magnetic field at the ^{89m}Y nuclear site. Here probably the transverse magnetic field—responsible for the ^{59}Co frequency pulling—plays the dominant role. Because of this additional field the precession of ^{89m}Y is more or less "synchronized" by the collective ^{59}Co precession frequency. Thus the homogeneous broadening is indirectly introduced via the ^{59}Co excitation. (ii) There exists a strong impurity-nuclei nuclear-magnon coupling originating from the SN interaction. For the resonance behavior the intrinsic magnetic hyperfine interaction of ^{89m}Y is no longer relevant; the excitation energy of the collective ^{89m}Y-nuclear-magnon state is strongly influenced by the nuclear-magnon dispersion relation. The homogeneous broadening is due to this coupling by which the transverse relaxation rate of ^{89m}Y becomes comparable to the corresponding rate of ^{59}Co.

Our experiments show that the SN interaction seems to act on impurity nuclei, even if the frequency difference between the impurity-nuclei hyperfine interaction and the $k = 0$ nuclear-magnon mode is larger than the SN width and the frequency pulling of the host. These results should stimulate theoretical investigations for the understanding of the impurity-nuclei nuclear-magnon interaction.

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