

Observation of Mössbauer Resonance Line Splitting Caused by Rabi Oscillations

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We present the first experimental observation of the Rabi splitting of Mössbauer resonance lines. A radio-frequency magnetic field is tuned to induce transitions between hyperfine sublevels of the Zeeman-split excited or ground state of ^{57}Fe nuclei. The line splitting corresponding to the Rabi flopping frequency is observed using a magnetically soft $^{57}\text{FeNi}$ absorber foil of composition with almost zero magnetostriction.

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A nuclear system exhibiting static hyperfine Zeeman splitting perturbed by a radio-frequency (rf) magnetic field is studied. The direction of the static magnetic field defines the quantization axis. The rf field is perpendicular to the static field and is tuned so as to induce transitions between the hyperfine sublevels of a Zeeman-split state. As a result of resonant magnetic excitation, the populations of these sublevels oscillate at the Rabi flopping frequency ω_R . In the classical description, the exponential decay of the gamma radiation field is modulated by a harmonic function with a frequency of $\omega_R/2$ [1, 2]. Recently, direct observations of the Rabi oscillations in the optical regime were reported [3, 4]. In the energy spectrum, the Rabi oscillations appear as a dynamic Stark splitting of the resonance lines when ω_R exceeds the experimental linewidth. This is the basic line shape to be seen in double resonance experiments under strong excitation.

Hack and Hamermesh [5] presented the first calculations of the double resonance for the 14.4-keV excited state of ^{57}Fe . Gabriel [6] and Mitin *et al.* [7, 8] treated the transverse rf magnetic field using the rotating wave approximation (RWA) and included the contributions from both nuclear states. A more general density-matrix formulation, not resorting to the RWA approach, has been developed by Salkola and Stenholm [9]. Up to now, despite the extensive theoretical work, there is no experimental evidence of the line splitting effect in Mössbauer spectroscopy, only line broadening has been observed [10, 11]. In fact, the very existence of such a line splitting effect in nuclear resonance has been questioned [12].

In this Letter we present the first experimental observation of the Rabi splitting in Mössbauer spectroscopy. The observed line shapes are uniquely and quantitatively explained by the theoretical description.

The obvious difficulty in the experiments with magnetic modulation of the ^{57}Fe Mössbauer resonance is associated with the natural linewidth of 1.1 MHz: The amplitude of the rf magnetic field at the nucleus must be several teslas for the line splitting to be visible. However, strong enough modulation can be produced by using magnetically soft ferromagnetic samples in which

the relatively weak external rf field is greatly enhanced [13]. With such samples, unfortunately, mechanically induced sidebands due to magnetostriction usually dominate the experimental data. We present experimental conditions where the influence of the mechanical vibrations is minimized. We used a sample of isotopically enriched $^{57}\text{Fe}_{18}\text{Ni}_{82}$ foil with a composition of almost vanishing magnetostriction and a specially constructed sample holder for elimination of mechanical resonances. The results indicate a transverse magnetic coupling of the rf field to the nuclear energy levels allowing observation of the line splitting. We emphasize that this is a new phenomenon in Mössbauer spectroscopy and that the observed line splitting effect is *definitely not* a mechanical sideband effect nor is it related to the collapse [14–16] of the static hyperfine structure.

The theory presented here is an application of the general density-matrix approach of Ref. [9]. If the rf field amplitude is low, this theory gives spectra which are in agreement with those obtained by Gabriel [6] and by Mitin, Makarov, and Polyakov [8]. The Hamiltonian of the system consists of the static and dynamic parts

$$\begin{aligned} \hat{H} &= \hat{H}_{\text{static}} + \hat{H}_{\text{dynamic}} \\ &= \hbar \begin{pmatrix} H_e & \mathcal{D} \\ \mathcal{D}^\dagger & H_g \end{pmatrix} + \hbar \begin{pmatrix} V_e & 0 \\ 0 & V_g \end{pmatrix} \cos(\Omega t). \end{aligned} \quad (1)$$

In the case of pure magnetic hyperfine interaction, the static level splitting can be characterized by $H_e = \Delta_0 + \Delta_e(B_0)I_{e,z}$ and $H_g = \Delta_0 + \Delta_g(B_0)I_{g,z}$, where Δ_0 includes the line shift parameter (usually the Doppler shift) and the Zeeman splitting corresponding to the static magnetic field B_0 is $\Delta_k(B_0) = -g_k\mu_N B_0/\hbar$ ($k = e, g$). The subscripts e and g refer to the excited and ground states, respectively, and $I_{k,z}$ is the spin operator. The $(2I_e + 1) \times (2I_g + 1)$ block matrix \mathcal{D} (with the indices n_e and n_g) describes the relative amplitudes of the gamma transitions between the hyperfine levels, i.e.,

$$\mathcal{D}_{n_e n_g} = \sum_{m_e m_g} a_{m_e}^{(n_e)} b_{m_g}^{(n_g)*} C(m_e, m_g) D_{m_e - m_g, \pm 1}^{(L)}(\phi\theta), \quad (2)$$

where $C(m_e, m_g)$ is the associated Clebsch-Gordan coefficient and $D_{m_e - m_g, \pm 1}^{(L)}$ is a rotation matrix element [17]. If the hyperfine structure consists of pure $|I_k, m_k\rangle$ states, the coefficients $a_{m_e}^{(n_e)}$ and $b_{m_g}^{(n_g)}$ are equal to the Kronecker-delta factor. The dynamic part of the Hamiltonian consists of periodic magnetic modulation at the frequency $\Omega/2\pi$. The modulation can be decomposed into a longitudinal and a transverse component, $V_k = \Delta_k(B_z^{\text{rf}})I_{k,z} + \Delta_k(B_x^{\text{rf}})I_{k,x}$, where B_z^{rf} and B_x^{rf} are the amplitudes of the rf field in the longitudinal z and in the transverse x direction.

The measurable quantity in Mössbauer experiments is the absorption A which can be calculated by [9]

$$A \propto \text{ImTr}\{\mathcal{D}^\dagger \hat{\rho}_{eg}\}, \quad (3)$$

where $\hat{\rho}_{eg}$ is the off-diagonal matrix block of the density-matrix operator. The Liouville equation describing the time dependence of the density matrix is

$$i\partial_t \hat{\rho}_{eg} = [H_e + V_e \cos(\Omega t)]\hat{\rho}_{eg} - \hat{\rho}_{eg}[H_g + V_g \cos(\Omega t)] + \mathcal{D}\hat{\rho}_{gg} - i\hat{\rho}_{eg}\Gamma/2, \quad (4)$$

where Γ is the linewidth of the excited state. In solving this equation, direct mathematical methods for complex differential equation systems can be utilized. On the other hand, a very convenient way is to search for the solution in the form $\hat{\rho}_{eg} = \sum_{k=-\infty}^{\infty} \rho_{eg}(k)e^{ik\Omega t}$, where the Fourier coefficients $\rho_{eg}(k)$ can be obtained with the matrix-continued-fraction techniques [18]. In calculating the absorption spectrum, only the coefficients $\rho_{eg}(0)$ are required.

Next we consider theoretically the effects of longitudinal and transverse rf fields on the ^{57}Fe Mössbauer resonance with $I_e = 3/2$, $I_g = 1/2$, $L = 1$ (the angular momentum carried by the photon), $B_0 = 30$ T, and $|\mathbf{B}^{\text{rf}}| = 8$ T. The linewidth Γ equals the natural linewidth and the angle between the propagation direction of the gamma rays and the quantization axis is $\theta = \pi/2$. In Figs. 1(a) and 1(b) the influence of a *longitudinal* rf field ($\mathbf{B}^{\text{rf}} \parallel \mathbf{z}$) is calculated at 10 MHz and at 23.6 MHz of which the latter value matches with the static level separation of the excited state. In addition to the six-line spectrum, well-known sidebands appear in these cases at the separations corresponding to the rf field frequency. The *transverse* case ($\mathbf{B}^{\text{rf}} \perp \mathbf{z}$) is studied in Figs. 1(c) and 1(d) for the resonance frequencies of the excited and the ground states, respectively. The spectra are characterized by the Rabi splitting corresponding to the multiplicity $2I_k + 1$ of the resonant state. When the applied rf field is in resonance with the ground state, the line splitting is given by $\Delta_g(B_x^{\text{rf}})/2$. A striking difference is observed between longitudinal and transverse modulation.

Our measurements were carried out in standard transmission geometry using a single line $^{57}\text{Co}:Rh$ source with an activity of 1.6 mCi. The external rf field was produced with a coil which was part of an LC resonance circuit.

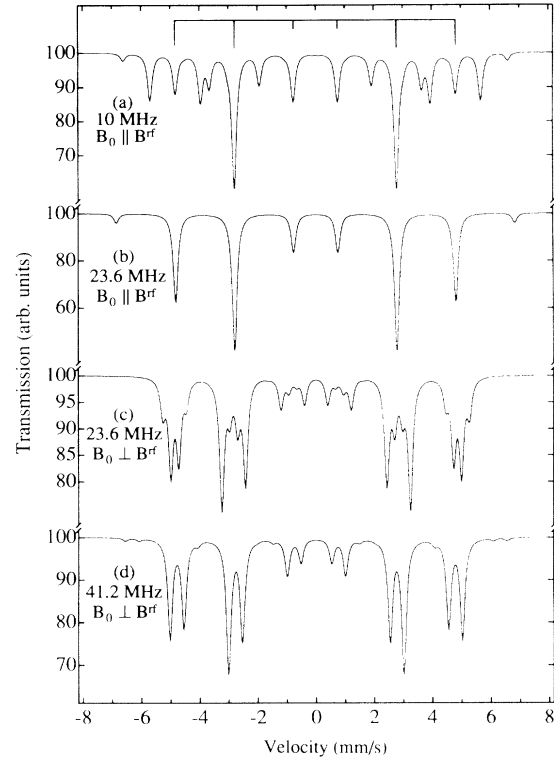


FIG. 1. Calculated Mössbauer spectra with rf excitation. The internal field is $B_0=30$ T and the rf field amplitude is 8 T. The line positions of the original six-line spectrum are indicated in the upper part of the figure. Spectrum (a) corresponds to 10-MHz longitudinal magnetic modulation. In spectrum (b) the rf field is applied longitudinally and in spectrum (c) transversely at the resonance frequency of the excited state (23.6 MHz). In spectrum (d) the transverse rf field is applied at the resonance frequency of the ground state (41.2 MHz).

Great care was taken to attach the FeNi foil rigidly to the plastic sample holder in order to prevent mechanical vibrations. Any heating effects were ruled out by cooling both the coil and the sample with water. The external rf field on the order of 10 mT was produced in the plane of the absorber foil perpendicular to the gamma rays. In addition, the rf field was cycled on and off at 1-s intervals and two spectra were accumulated during the measurement, one while the field was on and the other while the field was off. No external static magnetic field was applied. A more detailed description of the experimental setup will be reported in a separate publication.

The sample contained 81.98% nickel and 18.02% iron, enriched to 92.8% in ^{57}Fe . The thickness of the sample was $2.5 \mu\text{m}$ [19]. FeNi alloys form a homogeneous solid solution and each iron atom can have up to twelve iron atoms as nearest neighbors. Thus the samples exhibit a distribution of internal hyperfine fields at the iron nuclei. In the analysis of the spectra we used the approach de-

scribed by Rancourt and Ping [20] where the distribution of internal hyperfine magnetic fields can be characterized by two parameters: The field H_K with K iron nearest neighbors is given by the formula $H_K = H_L + H_T K$, where H_L and H_T are the local and transferred fields. The relative abundance of each nearest-neighbor configuration is taken from the binomial distribution. Since the iron concentration in our sample was rather low, no relaxation effects were considered [20].

The reference spectrum measured with no rf modulation is shown in Fig. 2(a). The schematic lines indicate the line positions and the relative intensities of the components used in the analysis of the spectra. Experimental results of Mössbauer measurements with magnetic rf excitation are shown in Figs. 2(b)–2(e). The spectra of Figs. 2(b) and 2(c) were measured with 15 W of rf

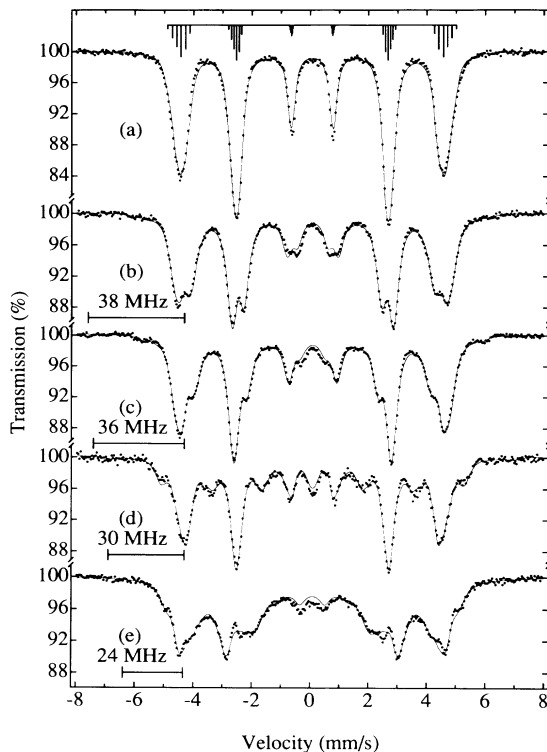


FIG. 2. Measured Mössbauer spectra with no rf modulation (a) and with transverse rf excitation (b)–(e). The line positions of the static reference spectrum due to the distribution of hyperfine fields are indicated. The rf field frequencies and rf powers are (b) 38 MHz, 15 W; (c) 36 MHz, 15 W; (d) 30 MHz, 11 W; and (e) 24 MHz, 15 W. The solid lines are least-squares fits and the velocity intervals corresponding to the rf field frequencies are indicated. Each of the six transitions of spectra (b) and (c) can be seen to split into two components corresponding to the NMR transitions between the sublevels of the ground state. The effects of nonresonant magnetic modulation can be seen in spectrum (d). In spectrum (e) the splitting to four components at the excited state resonance frequency remains largely unresolved.

power at the frequencies of 38 and 36 MHz, respectively. These values are close to the rf resonance frequency of the ground state. The spectrum 2(d) was measured with an 11-W rf field. The modulation frequency of 30 MHz is in the middle between the resonance frequencies of the ground and the excited states. Figure 2(e) corresponds to the rf field power of 15 W at the frequency of 24 MHz, close to the excited state resonance frequency.

Because of the Rabi oscillations driven by the transverse magnetic field, each of the six resonance lines in the ^{57}Fe Mössbauer spectrum splits into two or four components depending on whether the external field is in resonance with the ground or with the excited state sublevels. A small change of the frequency near the resonance results in large changes in the relative intensities of the split line components. Because of the distribution of internal hyperfine fields the measured intensity consists of components having different resonance frequencies. Yet, most of the split lines are well resolved in Fig. 2.

All the spectra of Fig. 2 were fitted (solid curves) by applying the above theory and by taking into account the distribution of hyperfine fields. The Rabi splitting is unambiguously demonstrated by the experimental spectra of Figs. 2(b) and 2(c). When the rf frequency is somewhat below the resonance frequency, the inner components of split lines have lower intensity [Fig. 2(c)]. Agreement between experimental and theoretical results is excellent, especially in the outer portions of the spectra. We also checked experimentally the validity of the theory for rf field frequencies away from either ground or excited state resonance. The spectrum in Fig. 2(d) corresponds to the rf field frequency of 30 MHz and additional lines appear as compared to the static six-line spectrum. By applying the resonance rf frequency of the excited state, a more complex spectrum results as each Mössbauer line splits into the four components. In Fig. 2(e) the four-fold splitting is not resolved due to the distribution of hyperfine fields. However, the general structure of the spectrum contains all the essential features of Fig. 1(c). It should be noted that these spectra can also be fitted using the RWA approaches of Refs. [6] and [8], but a slightly better agreement was found using the general theory of Ref. [9].

We also tried to analyze the spectra with the acoustic sideband model and with the sideband lines due to the hyperfine collapse phenomenon. It was clear that symmetrical sidebands cannot be used to explain these spectra since almost no lines corresponding to the separations of the rf field frequency appear outside the region of the original six-line spectrum. The presence of the collapse effect was ruled out by the absence of a strong line close to zero velocity. Since no sideband model could explain the measured spectra the only remaining possibility is a direct magnetic coupling of the rf field to the magnetic moments of the ^{57}Fe nuclei. This hypothesis was further corroborated by changes in line positions and line intensities corresponding to the rf-splitting effect as the

frequency of the external field was varied.

Since the magnetization of ferromagnetic domains remains in saturation under rf excitations, only rotation of the magnetization vector is possible. Thus the time-dependent rf magnetic field seen by the nucleus is always perpendicular to the effective static hyperfine field. Both transverse and longitudinal rf magnetic excitations were used to interpret the spectra. As expected, the effects of longitudinal rf excitations were negligible. The hyperfine field parameters obtained from the fits were $H_L = 24.3(6)$ T and $H_T = 1.3(1)$ T with the K values 0, . . . , 6 used in the fits. The amplitudes of the internal rf field varied between 4 and 8 T. These amplitudes slightly decreased both with increasing K and with increasing rf frequency. The isomer shift was $-0.18(4)$ mm/s relative to α -iron. In the analysis, the parameter ϕ in the rotation matrix element was averaged over 2π and only one θ angle was used. The fits of different spectra consistently gave a value $\theta=1.27(1)$ rad which corresponds to random polycrystalline foil [21].

In conclusion, the use of a magnetically soft FeNi sample enabled the rotation of the large internal hyperfine field causing an effective transverse magnetic modulation. In resonance with the static level splitting this field was successfully demonstrated to cause line splitting, indicating the presence of Rabi oscillations. From the analysis of the present spectra we found that the influence of mechanically induced sidebands on the results was negligible. The results are in excellent agreement with a strict theoretical model over a broad range of rf frequencies.

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