Softening of the phonon spectrum below the magnetic-phase-transition temperature

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Mössbauer studies of ^{57}Fe in $DyFe_2$ and YFe_2 at temperatures $0.5\,T_c \leq T \leq 1.1\,T_c$ were performed. Giant dips in the temperature dependence of the recoil-free fraction and the thermal shift were observed below T_c in $DyFe_2$. In YFe_2 a regular flat behavior was observed. These results are interpreted as evidence for the large influence of the spin fluctuations on the phonon spectrum through the huge magnon-phonon coupling present in $DyFe_2$ and absent in YFe_2.

It has recently been shown[1] that in magnetostrictive materials in the magnetic critical region spin fluctuations influence the phonon distribution and cause sharp changes in the ultrasonic attenuation coefficients. Other physical quantities which are sensitive enough to the phonon distribution should show similar phenomena. Two such quantities, which are measurable by the Mössbauer technique, are the Mössbauer recoil-free fraction[2] (f) and the second-order Doppler shift (δ).[3] In crystals with no magnon-phonon coupling the temperature dependences of $f(T)$ and $\delta(T)$ are smooth monotonic decreasing functions of temperature. However, if a strong magnon-phonon coupling is present, one may expect the spin fluctuations near the critical temperature (T_c) to influence the phonon distribution and affect both $f(T)$ and $\delta(T)$. We report here an experimental observation of this phenomenon.

The material chosen for this study was $DyFe_2$ because it has a large magnetostrictive coupling constant.[4] For comparison YFe_2 was chosen, as it is a crystallographically equivalent and magnetically similar material but shows very low magnetostriction. In practice we used samples of $Dy_{0.95}Tb_{0.05}Fe_2$ and $Y_{0.95}Ho_{0.05}Fe_2$ which both have the easy axis of magnetization along the [001] axis. This guarantees simple symmetric six-line spectra.[5]

Mössbauer studies were performed on ^{57}Fe in the temperature range 300–700 K. From these studies the Curie points were determined to be $T_c(Dy_{0.95}Tb_{0.05}Fe_2) = 638 \pm 3$ K, and $T_c(Y_{0.95}Ho_{0.05}Fe_2) = 554 \pm 3$ K.[6] Some of the Mössbauer spectra obtained are displayed in Fig. 1. One observes in Fig. 1 that for $T < T_c$ the spectra are composed of one symmetric well-resolved six-line pattern, though the quadrupole interaction is nonzero ($\frac{1}{4}eqQ$ is 0.15 mm/sec for $DyFe_2$ and 0.2 mm/sec for YFe_2 at 300 K). Such a pattern is expected when the magnetization is along the cubic [100] axis.[5] As the temperature is raised towards T_c the magnetic splitting decreases to zero, the quadrupole interaction also slightly decreases, but the spectra stay symmetric proving that the easy axis of magnetization is [100] up to T_c. The broad line observed at $1.02T_c$ is consistent with the presence of a quadrupole interaction. Above T_c the quadrupole interaction slightly increased. We determined T_c as the point where the spectrum reached its minimum width. The fact that this determination agrees so well with magnetization measurements[6] proves that the method is valid and that no changes occurred to our sample while the temperature was raised. The center shift of the spectra was determined by two methods, direct center of mass calculation and from the six-line least-square-fit

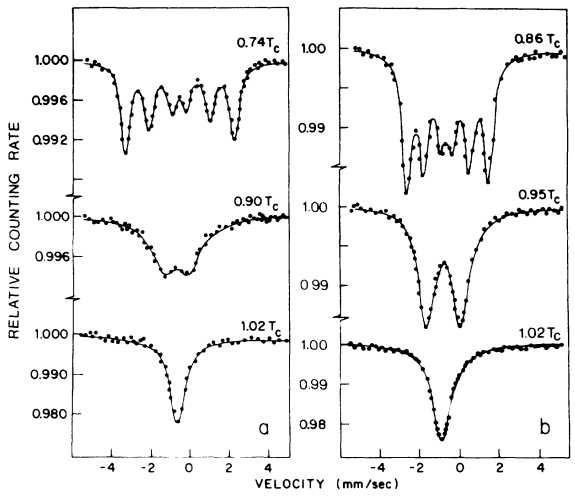

FIG. 1. Mössbauer spectra of ^{57}Fe in $Dy_{0.95}Tb_{0.05}Fe_2$ (26 mg/cm^2) (a), and $Y_{0.95}Ho_{0.05}Fe_2$ (31 mg/cm^2) (b). The solid curves represent theoretical best fit spectra composed of six Lorentzian absorption lines.

procedure. Close to T_c only the first method could be applied. This method is justified for symmetric spectra even if intensity saturation effects are present.

In addition to the ordinary Mössbauer spectra, the integrated (over the velocity range of the Mössbauer spectra) transmitted intensity through an opening angle of 1° was recorded by the method described in detail in a previous publication.[7] In Fig. 2 we present the integrated transmitted intensity (corrected for ~90% background[7]), 2(a), and the area under the Mössbauer absorption curve 2(b) as a function of temperature. We observe in Fig. 2(b) that in $DyFe_2$ below T_c a sharp decrease in resonant absorption occurs, accompanied of course by an increase in transmitted intensity, Fig. 2(a). In fact, Figs. 2(a) and 2(b) yield the same information, however, Fig. 2(a) is obtained with much less effort. Since the Mössbauer absorbers were not very thick (~0.2 mg/cm^2 ^{57}Fe) the area under the Mössbauer absorption curves is approximately proportional to the value of the

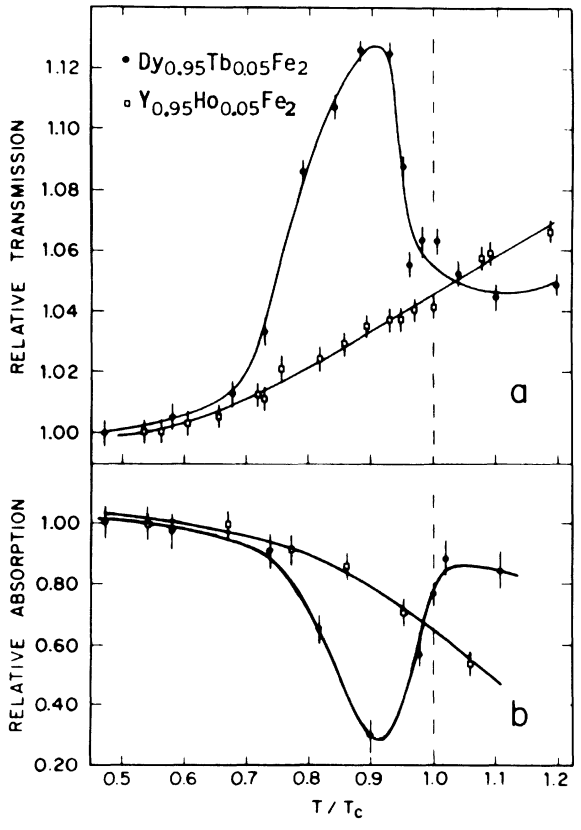

FIG. 2. (a) The transmitted intensity (normalized to room-temperature transmission) as a function of temperature. (b) The temperature dependence of the area under the Mössbauer absorption spectra, normalized to the area at room temperature.

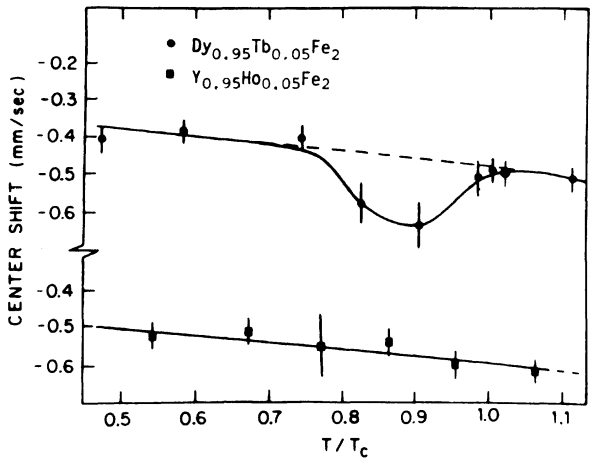

FIG. 3. The temperature dependence of the center of gravity of the Mössbauer spectra.

recoil-free fraction $f(T)$. A numerical estimate of the absolute value of $f(T)$ in $DyFe_2$ and its changes according to Fig. 2(b) show that $f(0.6T_c) \sim 0.6$ and $f(1.1T_c) \sim 0.5$, whereas $f(0.9T_c) \sim 0.2$. These results imply that in the region of $0.9T_c$, strong effective softening of the phonon spectrum occurs.[2] As observed in Figs. 1 and 2, the YFe_2 system shows no anomaly in the whole temperature range, leading us to believe that the observed phenomenon is to be associated with the large magnetostrictive coupling present in $DyFe_2$ and absent in YFe_2.

The large change in the phonon distribution in $DyFe_2$ should also influence the thermal shift $[\delta(T)]$.[3] This shift is experimentally determined by measuring the center of gravity of the Mössbauer spectrum. Our results show that in the region where phonon softening occurs the thermal shift does change drastically (Fig. 3). As expected, no anomaly in the thermal shift was observed for YFe_2 (Fig. 3).

In order to express the observed phonon softening in some quantitative manner let us present $f(T)$ and $\delta(T)$ in the Debye-model approximation, though we do not imply that this model should really describe the phonon distribution in the RFe_2 systems. In the Debye model approximation $f(T)$ and $\delta(T)$ can be expressed by simple closed form formulas

$$f(T) = \exp\left\{-\frac{6R}{k\Theta}\left[\frac{1}{4}+\left(\frac{T}{\Theta}\right)^2\int_0^{\Theta/T}\frac{x\,dx}{e^x-1}\right]\right\} \qquad (1)$$

and

$$\frac{\delta(T)}{E_0} = -\frac{9kT}{4Mc^2}\left[\frac{1}{4}\left(\frac{\Theta}{T}\right)+2\left(\frac{T}{\Theta}\right)^3\int_0^{\Theta/T}\frac{x^3dx}{e^x-1}\right], \qquad (2)$$

E_0 is the Mössbauer-transition γ energy, M is the mass of the Mössbauer nucleus, R is the recoil energy of a free nucleus, and Θ is the Debye temperature. One observes that $f(T)$ and $\delta(T)$ are indeed smooth monotonic decreasing functions of temperature, in particular for $T > \Theta$ one obtains

$$f(T) = \exp[-(6R/k\Theta)(T/\Theta)]$$

and

$$\delta/E_0 = -(3kT/2Mc^2)[1 + \tfrac{1}{20}(\Theta/T)^2] .$$

Using the Debye formula for $f(T)$ (assuming $\Theta \sim 300$ K) the decrease in f at $0.9T_c$ can be interpreted as a decrease in the effective Debye temperature by about 40%. The decrease in Θ is not consistent, according to Eq. (2), with the observed direction of the change in $\delta(T)$.

Thus we are led to the conclusion that our experimental observations of $\delta(T)$ and $f(T)$ cannot be explained in a consistent way within the Debye-model approximation. In fact, it was suggested[8] that the observed change in $\delta(T)$ can be a result of changes in the isomer shift produced by the rearrangement of conduction electrons when magnetic order sets in. However, it is also known[3] that the magnetostrictive coupling should introduce changes in the thermal shift below T_c. Thus the origin of the irregular behavior of $\delta(T)$ is not yet clear.

The irregular behavior of $f(T)$, $\delta(T)$, and $eq(T)Q$ in pure $TbFe_2$, $HoFe_2$, and $ErFe_2$ reported by Kimball *et al.*[8] agrees well with our observations. The change in the electric field gradient at T_c is probably due to the disappearance of the magnetostrictive distortion.

Though a detailed theory of the effects of the magnon-phonon coupling on $f(T)$ still does not exist we can understand the experimental decreases of $f(T)$ below T_c qualitatively. Below T_c the spin fluctuations coupled to the phonon bath introduce new channels to absorb the recoil energy of the Mössbauer nucleus. The density of the spin fluctuations decreases as T decreases and thus $f(T)$ increases again at low temperatures.

Our measurement of $f(T)$ in $DyFe_2$ and YFe_2 does indicate that in systems in which large magnetostriction is present effective phonon softening is expected below T_c.[9] It is obvious that this phenomenon can be studied also by measuring other physical quantities such as x-ray and neutron-diffraction line intensities, specific heat, sound velocity, and sound attenuation coefficients. However, the measurement of the temperature dependence of the Mössbauer recoil-free fraction is extremely simple if one uses the "integrated intensity" method suggested in the present paper. Our present results should also encourage theoreticians to work out a full theory of the magnon-phonon coupling effect on the temperature dependence of the Mössbauer recoil-free-fraction parameter.

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[9]It seems that this phenomenon has been observed also in the systems Fe_3C, FeF_3, and $NiFe$ (Ref. 7) but unfortunately erroneously interpreted. We now suspect that the angular dependence reported in Ref. 7 has no physical importance, it arises from the competition between absorption and scattering spectra, both of which are recorded under the conditions of that experiment.