ON EFFECT OF SPIN-DENSITY WAVE PARAMETERS ON SHAPE OF ^{119}Sn MÖSSBAUER SPECTRA

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Based on a simple numerical procedure the influence of various parameters characteristic of spin-density waves on the shape of ^{119}Sn Mössbauer spectra and the related histograms of the hyperfine field distributions is investigated.

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1. Introduction

Antiferromagnetism of chromium is constituted by the so-called spin-density waves (SDW's) [1]. According to Young and Sokoloff [2], the magnetic moment of chromium, $\mu(r)$, can be expressed as a series of odd-harmonics, μ_{2i-1}:

$$\mu(r) = \sum_{i=1}^{\infty} \mu_{2i-1} \sin[(2i-1)\alpha + \delta], \tag{1}$$

where $\alpha = \boldsymbol{Qr}$, $\boldsymbol{Q}$ being the wave and $\boldsymbol{r}$ — the position vector. The phase shift between the SDW and the lattice is given by δ. SDW's of chromium are incommensurate with the underlying lattice, i.e. $Q \approx 2\pi/a$, a being the lattice constant. If one introduces an incommensurability parameter, ϵ, one can write

$$Q = \frac{2\pi}{a}(1-\epsilon). \tag{2}$$

The periodicity of the SDW, Λ, can be defined as follows:

$$\Lambda = \frac{2\pi}{Q} = \frac{a}{(1-\epsilon)}. \tag{3}$$

It follows from Eq. (3) that $\Lambda \geq a$. In certain experiments, e.g. in the Mössbauer spectroscopy, the SDW is "seen" by probe nuclei occupying lattice sites. In such a case, the probe nuclei "see" the following periodicity of the SDW:

$$\Lambda_a = \frac{a}{\epsilon}. \tag{4}$$

In the case of chromium Λ_a lies between $\approx 20a$ at 4.2 K and $\approx 28a$ at 295 K [1].

If $\epsilon = 0$, the SDW is said to be commensurate with the lattice, which in terms of the periodicities means $\Lambda = a$ and $\Lambda_a = \infty$. However, one can also define another case of the commensurate structure, namely such that $\Lambda_a = na$, where n is a natural number.

The phenomenon of the SDW in chromium has been attracting a great interest of both theoreticians and experimentalists [1–3]. It presents an extremely interesting object for testing various models and theories concerning the magnetic and electric phase transitions for the former, and it can be successfully investigated by a variety of experimental methods, with the diffraction experiments in particular.

However, other experimental tools are also suitable for this purpose. They frequently constitute not only an alternative method, but in many cases they are complementary. One of them is the Mössbauer spectroscopy, whose potential has not been yet fully explored in the investigation of the issue, although already almost thirty years ago Street and Window demonstrated its ability in such studies [4]. About ten years ago Dubiel successfully applied this method to observe for the first time the spin-flip transition [5], and together with Le Caër they were able to determine the sign and the amplitude of the third-order harmonic of the SDW [6]. The authors also theoretically studied the influence of various parameters characteristic of the SDW on the shape of ^{119}Sn Mössbauer spectra [7].

In this paper the results of a further study of that kind based on a new numerical method we have developed for this purpose will be presented.

2. The way of the construction of the Mössbauer spectrum

For the sake of the symmetry, we consider a half-period of the SDW. First, for a chosen number of the harmonics of the SDW we calculate the shape of the resulting wave. Next we divide the half-period into N equidistant intervals, as

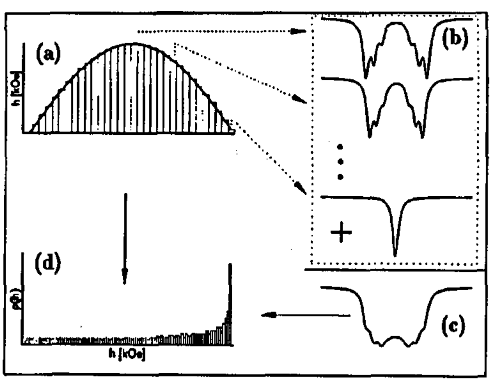

Fig. 1. Scheme of the construction of the Mössbauer spectrum from the given shape of the SDW. (a) The half-period of the SDW, (b) partial subspectra having the hf field proportional to the amplitude of the SDW in the given interval of its division, (c) the overall spectrum, (d) the histogram of the spin-density distribution.

schematically shown in Fig. 1a (for the incommensurate SDW, ISDW, N should be equal to infinity). For each of such intervals we calculate the amplitude of the wave and construct then a Mössbauer spectrum, which we shall call a partial spectrum, with the splitting proportional to the amplitude, i.e. we assume a linear relationship between the amplitude and the hyperfine field (spin-density) — see Fig. 1b. The overall spectrum is obtained by adding all the partial spectra — see Fig. 1c. The procedure enables also a construction of the histogram of the hf field distribution either directly from the SDW or from the final spectrum — see Fig. 1d.

3. Fundamental SDW, $h = H_1 \sin(\alpha + \delta)$

Theoretically, for a finite value of N the SDW is commensurate. However, as already shown in [7] for $h = 60 \sin \alpha$, if $N \geq 16$ the actual shape of the spectrum is indistinguishable from that characteristic of the ISDW. It is obvious that the critical value of N, N_c, i.e. such above which there is no measurable difference between the actual spectrum and the spectrum characteristic of the ISDW, will depend on the amplitude of the SDW, H_1. In order to show how N_c depends on H_1, we calculate the following difference parameter Δ:

$$\Delta = \sum_{i=1}^{M} [F(x_i, N) - F(x_i, 50]^2, \tag{5}$$

where M is a number of points constituting the spectrum (or number of channels),

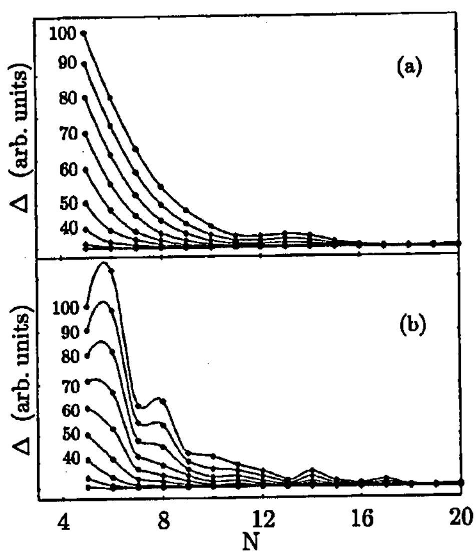

Fig. 2. The parameter Δ (see Eq. (5)) as a function of the modulation length, N, for (a) sinusoidal and (b) cosinusoidal SDW's. The values indicated stand for the amplitudes of the SDW's, H_1.

$F(x_i, N)$ is a number of counts in the i-th channel of the spectrum consisting of N partial spectra, $F(x_i, 50)$ is a number of counts in the i-th channel of the spectrum consisting of 50 partial spectra.

Figure 2 illustrates the quantity Δ as a function of N for the sinusoidal (a) and the cosinusoidal (b) modulation of the SDW for various values of H_1 shown. It is clear from that figure that $\Delta \approx 0$ for $N \geq 16$ even for the largest values of H_1. With decreasing value of H_1, the N_c shifts towards smaller values. In all the following calculations and simulations we use the value $N = 30$. In the examples of the spectra we will show it was also assumed that the lines are Lorentzian with the FWHM = 0.9 mm/s (which corresponds to the real width of a single line for ^{119}Sn Mössbauer isotope), and the relative amplitudes of the lines in the partial sextets are equal to 3:2:1.

4. Fundamental SDW, $h = H_1 \sin\alpha$

An example of the spectrum obtained by means of the above-described procedure for $h = |60 \sin\alpha|$ is shown in Fig. 3a, and the related histogram of the hf field distribution in Fig. 3b (in form of bars). The distribution obtained analytically is indicated in the same figure by a solid line.

In Fig. 4 the influence of the amplitude H_1 on the shape of ^{119}Sn spectra is visualized. The spectrum with $H_1 = 60$ kOe corresponds to the real spectrum of a single-crystal sample of Cr measured at 295K, and that with $H_1 = 100$ kOe to the one recorded at 4.2 K.

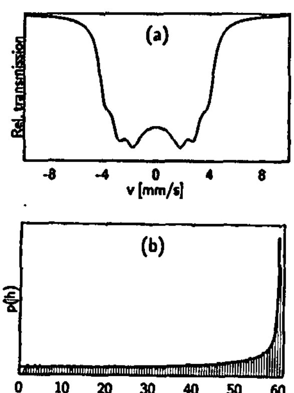

Fig.3

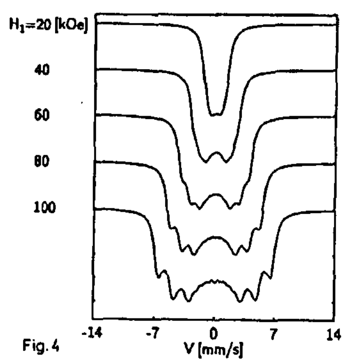

Fig.4

Fig. 3. Simulated ^{119}Sn Mössbauer spectrum for SDW's in case of $h = |60 \sin\alpha|$ (a) and related hf field distribution histogram (b) obtained numerically (bars) and analytically (solid line).

Fig. 4. Simulated ^{119}Sn Mössbauer spectra for SDW's in case of $h = |H_1 \sin\alpha|$. The numbers indicate H_1 values..

5. Higher-order harmonics, H_{2i-1}

5.1. Third-order harmonic, H_3

We shall consider the effect of the sign and the amplitude of H_3. This has not only purely theoretical reasons, as in a real sample of a single-crystal chromium $H_3 > 0$ and it amounts to $\approx 1.5\% H_1$ [6], and in polycrystalline samples of chromium $H_3 < 0$ and may reach $\approx 25\% H_1$ [8], depending on the size of grains.

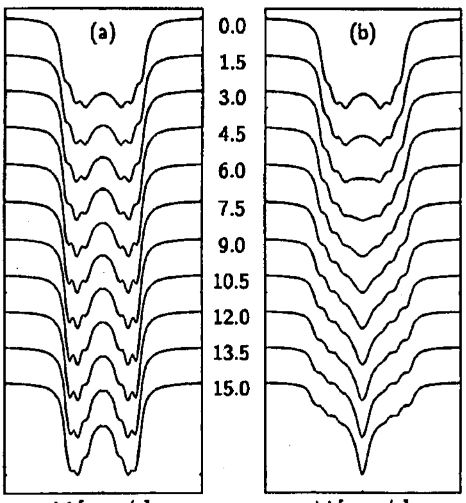

Fig. 5

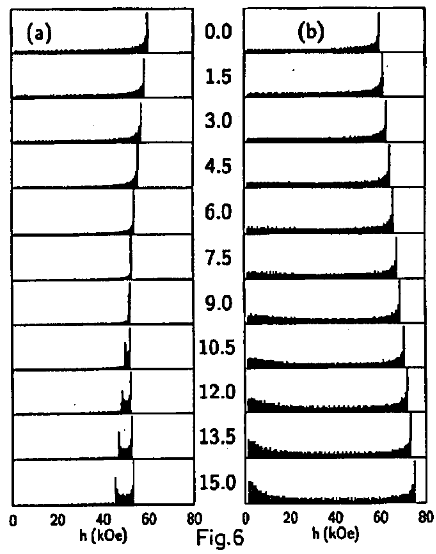

Fig.6

Fig. 5. Simulated ^{119}Sn Mössbauer spectra for SDW's in case of $h = |60 \sin\alpha + H_3 \sin 3\alpha|$ (a) and $h = |60 \sin\alpha - H_3 \sin 3\alpha|$ (b). The values indicated stand for the amplitudes of the H_3.

Fig. 6. The histograms of the hf field distributions derived from the spectra shown in Fig. 5 for $H_3 > 0$ (a) and $H_3 < 0$ (b). The labels correspond to those of Fig. 5.

A set of the spectra for $h = |60 \sin\alpha + H_3 \sin 3\alpha|$ is presented in Fig. 5 with $H_3 > 0$ (a) and $H_3 < 0$ (b) for various values of H_3 shown. The effect of both the sign and the magnitude of H_3 is obvious.

Related histograms of the hf field distributions are displayed in Fig. 6a and 6b, respectively. Here it can be seen that $H_3 < 0$ causes an increase in the maximum value of the hf field, $H_{\max}$, and, in addition, an appearance and a subsequent increase in the intensity of the zero field component. On the other hand, the presence of $H_3 > 0$ results first, in the decrease in $H_{\max}$, and next in its increase. Additionally, for $H_3 > 9$ kOe there is a second peak in the distribution, whose position shifts towards smaller values of h when H_3 increases.

In order to describe qualitatively the changes in the histograms due to the presence of H_3, we will analyze the behaviour of the function

$$h = \sin\alpha + R \sin 3\alpha \tag{6}$$

versus $R = H_3/H_1$. The function can be written as

$$h = (1 + 3R)\sin\alpha - 4R\sin^3\alpha. \tag{7}$$

The first derivative of the function (7), h', is equal to

$$h' = \cos\alpha(1 + 3R - 12R\sin^2\alpha). \tag{8}$$

It is equal to zero for the following two cases:
(i) when $\cos\alpha = 0$. In this case the maximum of the function h, $h_{\max}$, is given by

$$h_{\max} = 1 - R, \tag{9}$$

(ii) when $12R\sin^2\alpha = 1 + 3R$. In this case the maximum of h is given by

$$h_{\max} = \frac{2}{3}\sqrt{\frac{3R+1}{12R}}(3R+1), \tag{10}$$

if

$$\left(R \le -\frac{1}{3}\right) \cup \left(R \ge \frac{1}{9}\right). \tag{11}$$

The second derivative of h enables us to find its points of inflexion, which correspond to the further extreme of histograms. They are equal to

$$h_1 = 0 \tag{12}$$

and

$$h_2 = \frac{8}{9}\sqrt{\frac{27R+1}{36R}} \tag{13}$$

with

$$\left(R \le -\frac{1}{27}\right) \cup \left(R \ge \frac{1}{9}\right). \tag{14}$$

It should be also mentioned that h_2 always corresponds to the minimum of the distribution function, while h_1 corresponds to its minimum for $R > 0$ and to the maximum for $R < 0$.

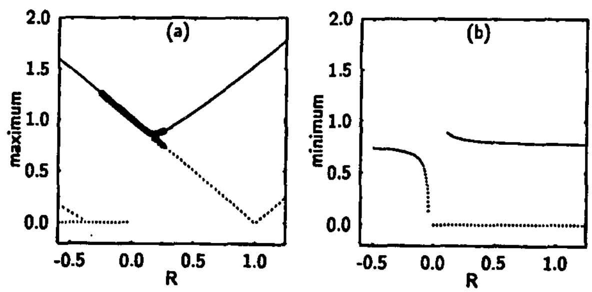

Fig. 7. The maxima (a) and the minima (b) in the distributions of the hf field versus the relative value of the third-order harmonic, $R = H_3/H_1$ for the SDW given by $h = |\sin\alpha + R\sin 3\alpha|$. The circles and triangles indicate the corresponding values found numerically.

Figure 7 illustrates the above-described relations showing the maxima (a) and the minima (b) as a function of R. Also in Fig. 7a one can see several maxima found numerically. They agree perfectly well with those found analytically which proves that our numerical procedure works correctly.

5.2. Fifth-order harmonic, H_5

To illustrate the effect of H_5 on the shape of the spectra, we show in Fig. 8 examples of such spectra calculated for the SDW described by $h = |H_1 \sin\alpha + H_3 \sin 3\alpha + H_5 \sin 5\alpha|$ with various values of H_1, H_3 and H_5. It is clear from the figure that the spectrum is sensitive both to the sign and to the magnitude of H_5.

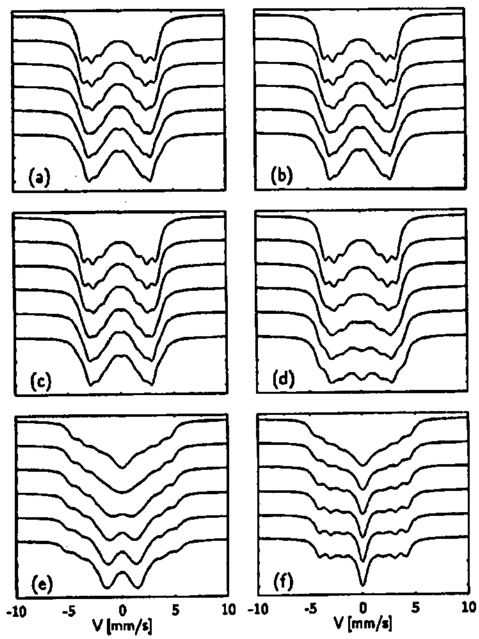

Fig. 8. Examples of the change of the shape of the SDW due to the presence of the 5-th order harmonic (downwards, from 0 to 10, step 2) for: (a) $h = |60 \sin\alpha + H_5 \sin 5\alpha|$, (b) $h = |60 \sin\alpha - H_5 \sin 5\alpha|$, (c) $h = |60 \sin\alpha + 10 \sin 3\alpha + H_5 \sin 5\alpha|$, (d) $h = |60 \sin\alpha + 10 \sin 3\alpha - H_5 \sin 5\alpha|$, (e) $h = |60 \sin\alpha - 10 \sin 3\alpha + H_5 \sin 5\alpha|$, (f) $h = |60 \sin\alpha - 10 \sin 3\alpha - H_5 \sin 5\alpha|$.

As already mentioned, a consideration of the higher-order harmonics of the SDW is not only of a theoretical meaning, but of a practical one, too. The reader is referred to our recent paper [8], where we give an evidence that the Mössbauer spectra of polycrystalline samples of chromium can be well described provided the higher-order harmonics are taken into account.

6. Summary

We have outlined a simple, numerical procedure that permits a simulation of ^{119}Sn Mössbauer spectra for sinusoidally modulated SDW's. It enables to investigate the influence of various parameters characteristic of the SDW such as the sign and the magnitude of the higher-order harmonics, as well as the phase shift between the SDW and the lattice on the shape of the spectra and the related histograms of the hf field distributions. The procedure in its iterative version has been successfully applied to analyze various ^{119}Sn Mössbauer spectra recorded on various samples of chromium [8].

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