

Influence of vanadium on spin- and charge-density waves in chromium

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(Received 17 April 1995; revised manuscript received 5 July 1995)

^{119}Sn Mössbauer spectroscopy is applied to study the influence of vanadium on spin- and charge-density waves in single-crystal and polycrystalline samples of chromium at 4.2 and 295 K. It has been found that V doping drastically quenches spin-density waves (SDW's). In single crystals the amplitude of the SDW, which was assumed to be proportional to the maximum hyperfine (hf) field, H_{max} , and the average hf field, $\langle H \rangle$ decrease linearly with the concentration (x) of V. The former at the rate of 26.2 kOe/at. % and the latter at 18.7 kOe/at. %. The normalized decrease of both quantities, $N = H_{\text{max}}$ or $\langle H \rangle$, was found to be the same, $(dN/N)/dx = -0.3/\text{at. \%}$. It agrees well with the normalized decrease of the Néel temperature, the average magnetic moment, and the incommensurability wave vector. This strongly supports the claim that the spectral parameters viz. H_{max} and $\langle H \rangle$ are directly related to the density of electrons at the Fermi surface and further supports the canonical model for SDW antiferromagnetism of chromium. Enhanced values of all spectral parameters characteristic of the SDW's and charge-density waves (CDW's), except the $\langle H \rangle$ values, have been determined in the polycrystalline samples. The enhancement is in part due to the interactions of SDW's and CDW's with grain boundaries, and partly to the internal strain. Residual SDW's have been revealed to exist in these samples even at 295 K.

I. INTRODUCTION

The magnetic structure of chromium is due to spin-density waves (SDW's). The imperfect nesting of electron and hole Fermi surfaces was recognized by Overhauser as their origin.¹ SDW's of chromium and its alloys have been attracting the interest of the experimentalist because of the great variety of phenomena associated with the SDW and the possibility of their investigation by means of numerous experimental techniques.² The theorist has been motivated to study Cr and Cr alloys because they constitute a special case of alloy systems on which various theoretical models depicting electric and magnetic structures and phase transitions related to the nesting Fermi surface can be tested and compared. Similarities between spin fluctuations in the paramagnetic phase of Cr,³ on one side, and those in the high-temperature superconducting cuprates,⁴⁻⁶ on the other, have additionally reinforced a further interest in the investigation of the itinerant nature of magnetism in these materials with the hope of helping us to understand both better. The canonical model for SDW antiferromagnetism in Cr alloy systems, which incorporates nesting Fermi surfaces and whose comprehensive description can be found in the review by Fawcett,² provides a satisfactory explanation of several physical properties of the systems. As a result of experimental data found for CrV (V acts as an electron acceptor) and CrMn (Mn acts as an electron donor) alloy systems, and, in particular, the linear decrease of the Néel temperature, T_N , the SDW incommensurability wave vector, Q^* and the SDW amplitude with V concentration,² the use of the paramagnetic alloy Cr_{95}V_5 as a reference material⁷⁻⁹ has served to confirm the essential features of the model. The successful interpre-

tation of this and other experimental data in favor of the canonical model by theoretical calculations of Machida and Fujita,¹⁰ who used an exact solution of the canonical model, has further supported it.

On the other hand, however, evidence was found that this ideal dopant (vanadium) produces a strong effect on several physical properties of chromium in the vicinity of T_N , and especially in the paramagnetic phase. In particular, at low concentrations V doping suppresses spin fluctuations,³ lowers the resistivity,¹¹ strongly reduces the inelastic neutron scattering,^{5,6} and destroys the first-order transition from the paramagnetic into the magnetic state.¹² This data requires a further development of the model in order to explain this rather unexpected behavior. The effect of impurities, and in particular that of vanadium, on the magnetic structure and the phase diagram of chromium has recently been theoretically studied by Fishman and Liu in terms of the mean-field free energy.¹³ The authors emphasize the need for additional experimental investigations of the CrV system in order to enlarge a basis on which this and other models can be tested and compared. In this respect the Mössbauer spectroscopy, and in particular at ^{119}Sn nuclei embedded in Cr V, seems to be an adequate technique to study the issue.¹⁴ Its main advantage lies not only in its high intrinsic resolution, but first of all in the fact that it supplies information on the SDW and the concomitant charge-density wave (CDW) simultaneously.

In this paper results obtained by means of such spectroscopy on single-crystal and polycrystalline samples of $\text{Cr}_{100-x}\text{V}_x$ alloys, for $0 \leq x \leq 5.3$, measured at 4.2 and 295 K are presented and discussed.

TABLE I. Chemical composition of the investigated samples of $(\text{Cr}_{100-x}\text{V}_x)_{100-y}\text{Sn}_y$.

Sample	Single	Single	Single	Poly	Poly	Poly
x [at. %]	0.5	2.5	5.0	0.6	2.5	5.3
y [at. %]	0.16	0.15	0.16	0.6	0.5	0.6

II. EXPERIMENTAL

A. Sample preparation and chemical composition

Single-crystal samples used in the present measurements were prepared from pieces donated by E. Fawcett. Thick platelets ($\sim 500 \mu\text{m}$) cut from the original crystal were first mechanically and then electrolytically polished down to the final thickness of $\sim 100 \mu\text{m}$. These in turn were covered on both sides with a thin layer of Sn enriched to $\sim 93\%$ in the ^{119}Sn isotope by sputtering, and subsequently annealed for 6 h at 1600 K in a protective atmosphere of Ar. By this procedure the probe nuclei have been diffused into the CrV matrices.

Polycrystalline samples of CrV alloys were prepared by melting appropriate amounts of 4N-pure Cr, 5N-pure V, and 3N-pure Sn, the latter enriched to $\sim 93\%$ in the ^{119}Sn isotope. The melting process was carried out in an arc-furnace filled with a protective Ar atmosphere, and it was repeated three times for each ingot to ensure a better homogeneity. The microstructure was observed by light microscopy and the average size of the grains determined by a lineal method was found to be $\sim 65 \mu\text{m}$.

The chemical composition of the polycrystalline samples was determined by atom emission spectroscopy. The concentration of V in the single crystals is as given by Fawcett, and the content of Sn was determined by microprobe analysis. The compositions of all the samples studied are presented in Table I.

B. Measurement of the Mössbauer spectra

^{119}Sn Mössbauer spectra of the investigated samples were registered in a transmission geometry using a standard spectrometer and a sinusoidal drive. Monoenergetic γ rays of 23.8 keV energy were emitted by a $\text{Ca}^{119\text{m}}\text{SnO}_3$ source kept at 4.2 or 295 K, respectively.

III. ANALYSIS OF THE SPECTRA

The spectra having a structure having more than a single line were analyzed in terms of higher-order harmonics of SDW's and the concomitant CDW's. Theoretically, SDW's can be described by a series of odd harmonics, H_{2i-1} :¹⁵

$$\text{SDW} = \sum_{i=1}^{\infty} H_{2i-1} \sin[(2i-1)\mathbf{Q}\cdot\mathbf{r} + \delta_s], \quad (1)$$

where \mathbf{Q} is the wave and \mathbf{r} the position vector, while δ_s is the phase shift between the SDW and the lattice.

SDW's through the spin-phonon interaction create charge-density waves (CDW's), which, in turn, can be expressed in terms of even-harmonics, I_{2i} :²

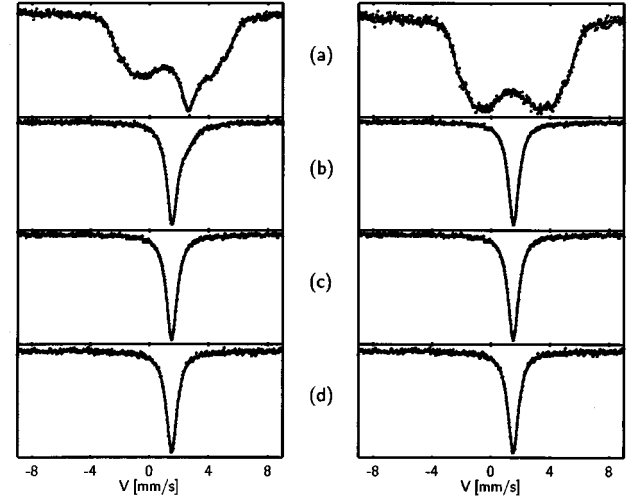


FIG. 1. ^{119}Sn Mössbauer spectra registered at 295 K on a single-crystal samples of a pure Cr (a) and $\text{Cr}_{100-x}\text{V}_x$ alloys with $x=0.5$ (b), $x=2.5$ (c), and $x=5.0$ (d) as measured (left-hand side) and after the removal of a single line due to the undissolved β -Sn (right-hand side). Solid lines represent the best-fit spectra.

$$\text{CDW} = \sum_{i=0}^{\infty} I_{2i} \sin(2i\mathbf{Q}\cdot\mathbf{r} + \delta), \quad (2)$$

δ being the phase shift between the SDW and the CDW.

Taking into account Eqs. (1) and (2) an iterative procedure for fitting the spectra has been developed and tested. Its main features are as follows:

(i) for a given set of harmonics of the SDW and the CDW a resulting wave is constructed. Its half-period (for the SDW) and the full period (for the CDW) is divided into n equidistant intervals, and for each of them a sextet is constructed with the splitting, H proportional to the amplitude of the wave in that interval and with the isomer shift, I proportional to the amplitude of the CDW in that interval.

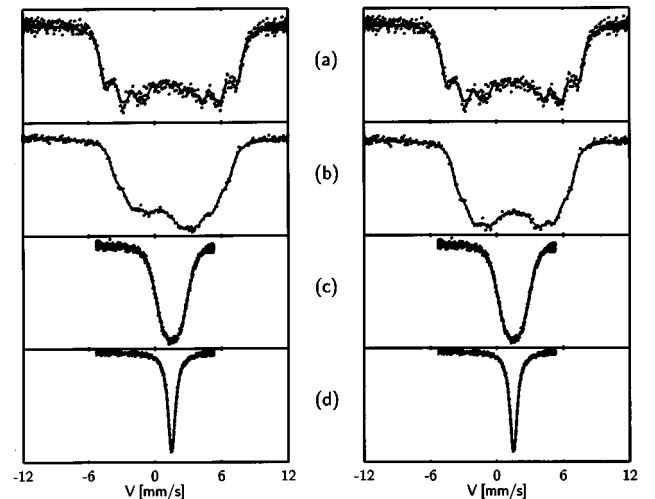


FIG. 2. The same as in Fig. 1 but measured at 4.2 K.

TABLE II. The best-fit spectral parameters obtained for single-crystal samples of $\text{Cr}_{100-x}\text{V}_x$ alloys by means of the method described in Ref. 16. H values are in kOe and I values in mm/s (I_0 being the average isomer shift).

	$T=4.2$ K	$T=4.2$ K	$T=4.2$ K	$T=295$ K
x	0	0.5	2.5	0
H_1	93.6(5)	75.5(9)	21.3(5)	57.1(5)
H_3	2.4(5)	0(1)	-2.2(3)	0.8(4)
H_5	-0.6(4)	1.0(5)	1.1(5)	0.6(3)
H_7	-0.2(4)	-1.3(3)	-0.9(7)	-0.5(4)
H_{\max}	91.1	79.1	25.5	57.4
$\langle H \rangle$	60.0	48.1	13.2	36.6
I_2	0.00(3)	0.00(2)	0.00(2)	0.00(2)
I_4	0.01(3)	0.01(3)	0.00(3)	0.03(2)
I_6	0.00(2)	0.00(3)	0.00(4)	0.02(2)
I_8	0.00(3)	0.00(3)	0.00(7)	0.001(2)
I_0	1.48	1.54	1.50	1.56

(ii) The overall spectrum is built up by a superposition of n subspectra obtained in the way described under (i).

For more information and details the reader is referred to Ref. 16.

IV. THE RESULTS AND THEIR DISCUSSION

A. Single-crystal samples

^{119}Sn Mössbauer registered at 295 K for a pure (110) face Cr and CrV alloys are presented in Fig. 1 (left-hand side, as measured, and right-hand side, after the line due to undissolved β -Sn has been removed). As it can be seen, all the spectra of CrV alloys consist of a single line only, which means that the samples are in the paramagnetic phase at this temperature. This observation agrees with the phase diagram of the system.¹⁷ The spectral parameters of the CrV spectra are the same within the error limits, irrespective of their composition. The full width of the line at half maximum,

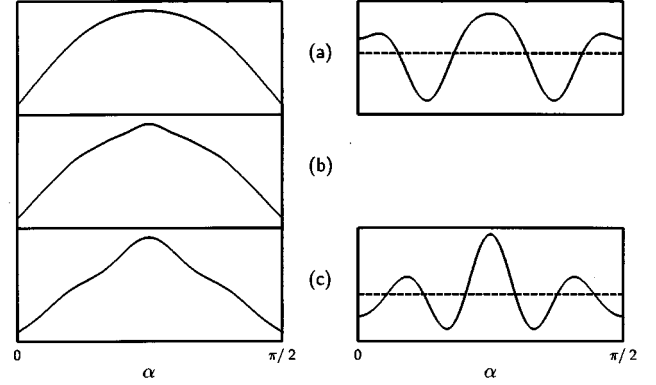


FIG. 3. The shapes of the SDW (left-hand side) and the CDW (right-hand side) derived from the spectra shown in Fig. 2. The labeling (a) through (c) corresponds with that in Fig. 2. The broken line indicates the average value of the isomer shift, I_0 ($\alpha=\mathbf{Q}\cdot\mathbf{r}$).

$\Gamma=0.96$ mm/s, and the isomer shift, $I=1.54$ mm/s. In the spectrum of the least-concentrated sample an additional line, indicated by an arrow, can be seen. It has been identified as due to β -Sn. More structure, hence more information, can be seen in the spectra registered at 4.2 K shown in Fig. 2. They are also displayed as measured (left-hand side), and after the removal of the line due to undissolved β -Sn (right-hand side). The spectrum of the Cr 0.5 at. % V sample is quite asymmetric. A possible reason for the asymmetry could be either a presence of the CDW—see Ref. 16, or a precipitation of a Sn-containing foreign phase. The former reason can be excluded as unlikely. In order to account for the observed asymmetry, the concomitant CDW should have its amplitude as large as 0.25 mm/s, which is not physical in this case. On the other hand, a light microscopic micrograph has revealed an existence of a foreign object within the matrix. We could not remove it either by etching or electropolishing. Its presence in the spectrum could be accounted for by a single line with the isomer shift of $I=2.55$ mm/s and $\Gamma=1.6$ mm/s. The other two spectra of CrV are free of undissolved tin, and the

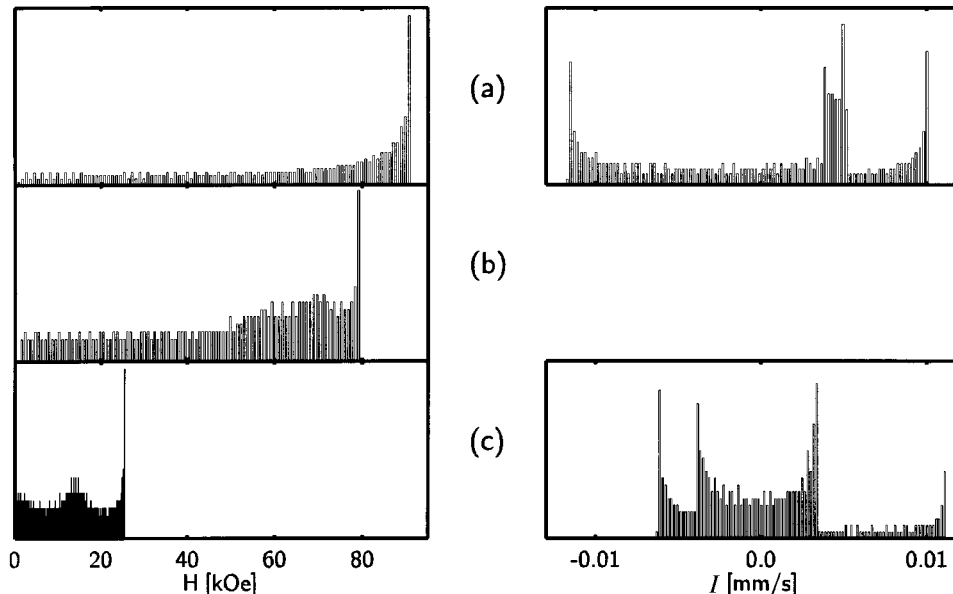


FIG. 4. The histograms of the hf field (left-hand side) and the isomer-shift (right-hand side) distributions obtained from the spectra shown in Fig. 2 for $x=0$ (a), $x=0.5$ (b), and $x=2.5$ (c).

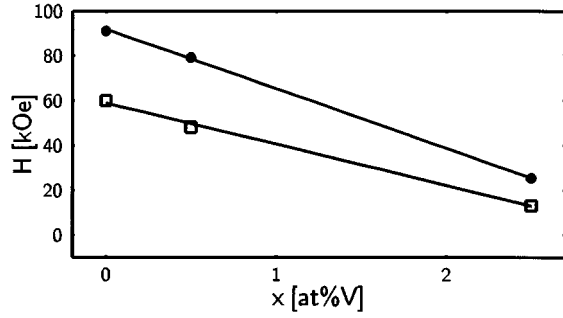


FIG. 5. The maximum hf field, H_{\max} (●) and the average hf field, $\langle H \rangle$ (□) as a function of x for the single crystals of $\text{Cr}_{100-x}\text{V}_x$ alloys measured at 4.2 K. The straight lines represent the best fits to the data.

spectrum of the most-concentrated sample can be fitted as a single line with the same Γ value as at 295 K. The spectrum of the least-concentrated sample was analyzed assuming only SDW's were present (the additional line due to the foreign phase, with *a priori* unknown spectral parameters, made it impossible to fit the spectrum in terms of coexisting SDW's and CDW's. The spectrum of the Cr 2.5 at. % V sample was fitted assuming both SDW's and CDW's are responsible for its shape. The best-fit parameters are displayed in Table II, while Figs. 3 and 4 show the shapes of the waves and the related histograms of the hyperfine field (spin-density) and the isomer shift (charge-density) distributions, respectively. The corresponding data obtained for the pure Cr has been included for comparison.

It follows from Fig. 3 and Table II that V doping strongly reduces the maximum value of the hf field, H_{\max} (hence the amplitude of the SDW). As illustrated in Fig. 5, H_{\max} decreases linearly with x at the rate of 26.2 kOe/at. %. The same figure shows that the average hf field, $\langle H \rangle$ also decreases linearly with x with the slope of 18.7 kOe/at. %. As mentioned in the Introduction, other physical quantities characteristic of the SDW, viz. T_N , $Q^* = -2\pi\epsilon/a$ ($\epsilon \approx 0.04$ being the incommensurability parameter and a the lattice constant) and the average magnetic moment $\langle \mu \rangle$ exhibit the linear decrease with x . In order to quantitatively compare the rate of the decrease of these quantities with the rate of decrease of H_{\max} and $\langle H \rangle$, we calculate the rate of decrease of the normalized quantities, $R = (dN/N)/dx$ for each of them ($N = T_N, Q^*, \langle \mu \rangle, H_{\max}$, and $\langle H \rangle$). The R values obtained in this way are displayed in Table III. It can be seen that within $\pm 10\%$, $R = \text{const}$. This means that all the quantities under consideration have the same relative change per at. % V embedded into the Cr matrix. In the light of the canonical model it means also that the spectral parameters H_{\max} and $\langle H \rangle$ have

TABLE III. Normalized rate of decrease, R of the average magnetic moment, $\langle \mu \rangle$, the Néel temperature, T_N , the incommensurability wave vector, Q^* , the maximum, H_{\max} and the average hf field, $\langle H \rangle$ for single-crystal $\text{Cr}_{100-x}\text{V}_x$ alloys. (+) calculated according to the data from Ref. 17.

	$\langle \mu \rangle$	T_N	Q^*	H_{\max}	$\langle H \rangle$
R	-0.30^+	-0.27^+	-0.26^+	-0.29	-0.31

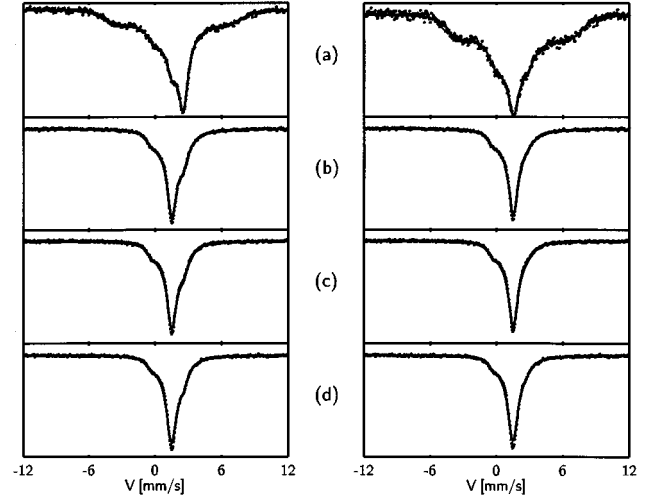


FIG. 6. ^{119}Sn Mössbauer spectra of polycrystalline samples of pure Cr (a) and its $\text{Cr}_{100-x}\text{V}_x$ alloys with $x=0.6$ (b), $x=2.5$ (c), and $x=5.3$ (d) registered at 295 K as measured (left-hand side) and after the removal of the line due to the undissolved $\beta\text{-Sn}$ (right-hand side). Solid lines stand for the best-fit spectra.

the same relation to perturbations of the density of states at the Fermi surface as T_N , Q^* , and $\langle \mu \rangle$. From the viewpoint of the experimental method used in the present study the above result qualifies it as an adequate method in the investigation of the SDW of chromium. One can regard H_{\max} as a good measure of the amplitude of the SDW and $\langle H \rangle$ as a good measure of the average amplitude of the SDW. Finally, one can also conclude that doping with V quenches the SDW as a whole, i.e., without changing its shape.

Concerning now the results obtained for the Cr 2.5 at. % V sample, an enhanced value of the third-order harmonic of the SDW, $H_3 = -2.2(3)$ kOe, i.e., $\sim 10\%$ of H_1 has been found. It differs in the magnitude and in the sign from its value found for a pure Cr. Such effect of an enhancement and the change of the sign have been revealed to be indicative of polycrystalline samples and grain-boundary effects.¹⁸ Why does one observe such an effect here in the single-crystal sample? The Laue pattern of the sample gave evidence that the crystal was not perfect which may, at least partly, be responsible for the higher relative value of H_3 and its reverse, negative sign.

Finally, as far as the CDW is concerned, one can see for the Cr 2.5 at. % V sample that its amplitude is, within error limit, equal to zero. This reflects the fact that the spectrum is symmetric and has no structure. This result may indicate that V doping also suppresses the CDW.

B. Polycrystalline samples

^{119}Sn Mössbauer spectra registered at 295 K on these samples are displayed in Fig. 6 together with a spectrum of a pure polycrystalline Cr. As for the single crystals, the left-hand side of the figure shows the spectra as measured, and the right-hand side presents the spectra with the removed line due to undissolved $\beta\text{-Sn}$. One can readily see that the addition of V drastically affects the shape of the spectrum, which however, does not depend on the V concentration. In contrast

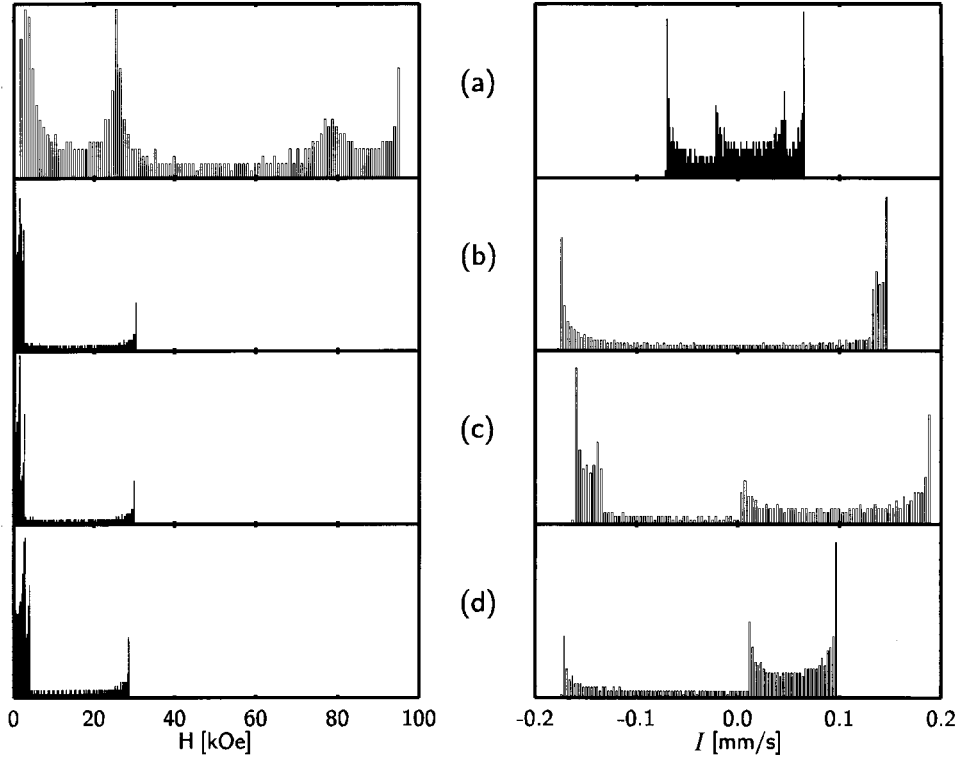


FIG. 7. The histograms of the hf field (left-hand side) and of the isomer shift (right-hand side) distributions derived from the spectra shown in Fig. 6. The labels (a) throughout (d) correspond with those in Fig. 6.

to the spectra registered on the single-crystal samples, see Fig. 1, those measured on polycrystalline samples show some residual splitting. This can be seen clearly in the histograms of the hf field distributions that are shown in Fig. 7. The spectra were analyzed in terms of the higher-order harmonics of the SDW and coexisting CDW. The best-fit spectra obtained are presented in form of solid lines in Fig. 6, and the best-fit spectral parameters derived from the spectra are displayed in Table IV. One can note that their values essentially do not depend on the concentration of V. There is merely a slight decrease of H_{\max} from 30.4 kOe for the least- to 28.7 kOe for the most-concentrated sample. The average hf field, $\langle H \rangle$ decreases from 8.2 to 6.3 kOe, respectively. The characteristic feature of the parameters is enhanced values of the higher-order harmonics of both SDW's and CDW's. They follow from the fact that the samples are polycrystalline, and

the interaction with grain boundaries obviously causes the enhancement.¹⁸ Before the results obtained from the spectra measured at 4.2 K will be presented and discussed, one should mention at this point that the residual SDW's and CDW's seen in our polycrystalline samples at 295 K very likely originate from the internal strain that exists in the samples. The strain has been generated by filing the ingots of the sample to prepare samples for the measurements of the Mössbauer spectra. It is well known that strain dramatically increases T_N of pure Cr,² i.e., it extends the temperature range of the existence of the SDW. In the light of the R values shown in Table III, the increase of T_N should be accompanied by an appropriate increase of other quantities characteristic of the SDW, and in particular of H_{\max} .

Another argument in favor of the picture of the strain-supported waves is as follows: The spectra shown in Fig. 6

TABLE IV. The best-fit spectral parameters as obtained for polycrystalline samples of $\text{Cr}_{100-x}\text{V}_x$ alloys by means of the method outlined in Ref. 16. H values are in kOe and I values in mm/s.

	4.2 K	4.2 K	4.2 K	295 K	295 K	295 K	295 K
x	0.6	2.5	5.3	0	0.6	2.5	5.3
H_1	62(8)	25(2)	17(2)	69(2)	15(1)	14(4)	12(7)
H_3	-10(4)	-4(5)	-12(4)	-21(1)	-10(2)	-11(4)	-10(9)
H_5	8(4)	3(6)	6(9)	5(2)	3(1)	5(2)	7(3)
H_7	0(6)	-1(3)	-2(9)	0(1)	0.5(9)	0(2)	
H_{\max}	85.8	33.7	37.5	30.4	30.9	30.0	28.7
$\langle H \rangle$	38.3	15.3	9.1	8.2	8.0	7.5	6.3
I_2	-0.1(3)	0.2(5)	0.0(3)	0.18(3)	-0.2(8)	0.1(2)	0.1(2)
I_4	-0.1(2)	-0.1(2)	0.1(4)	-0.02(5)	0.02(5)	-0.1(3)	-0.1(4)
I_6	0.0(2)	-0.1(3)	0.2(3)	0.02(9)	0.05(6)	-0.1(1)	
I_8	0.0(2)	0.0(3)	0.1(3)		0.01(7)		
I_0	1.41	1.47	1.50	1.41	1.49	1.42	1.45

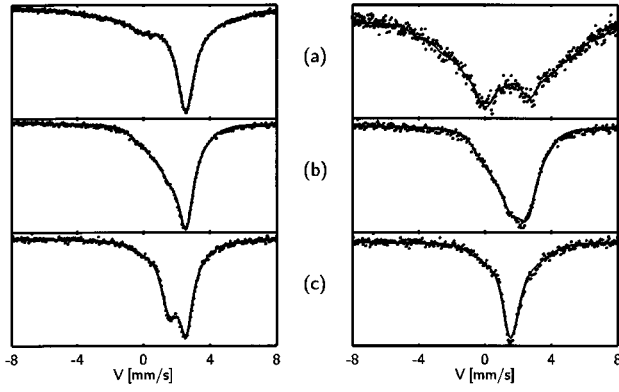


FIG. 8. The same as in Fig. 6 but measured at 4.2 K.

could not be well fitted with the assumption they originate from SDW's and CDW's only. It was also necessary to include a single line. This may indicate that SDW's do not exist in the entire volume of the polycrystalline samples at 295 K, but only within those regions where the strain is large enough. Otherwise, the remaining volume is paramagnetic.

The spectra of the studied polycrystalline samples measured at 4.2 K are presented in Fig. 8. They were also analyzed in terms of higher-order harmonics of SDW's and coexisting CDW's. The best-fit spectra obtained in this way are shown as solid lines in Fig. 8, and the best-fit spectral parameters are included in Table IV. The related histograms of the hf field and the isomer shift distributions are visualized in Fig. 9. As can be seen the decrease of temperature has resulted in the increased contribution of the line due to the undissolved β -Sn, which reflects the fact that the recoilless fraction for β -Sn is about five times larger at 4.2 K than at 295 K. Concerning the spectral parameters, the decrease of temperature has caused an increase of H_{\max} and $\langle H \rangle$ for all three samples. However, the magnitude of the increase depends on the sample. In particular, for the least-concentrated sample H_{\max} has increased by the factor 2.8 and $\langle H \rangle$ by 4.7. For the medium- and the most-concentrated samples the increase lies between a factor of 1.1 and 2. As far as the shape

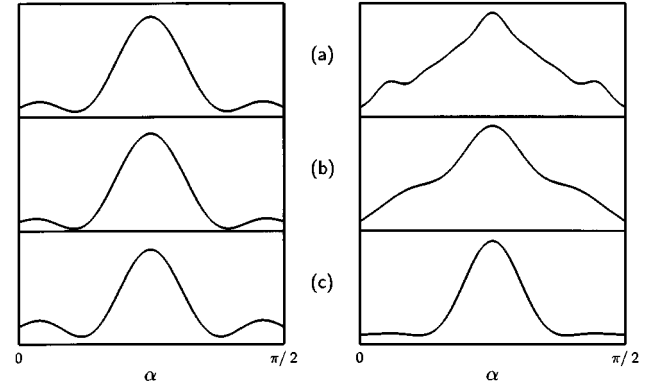


FIG. 10. The shapes of the SDW obtained from the spectra illustrated in Fig. 6 (left-hand side) and in Fig. 8 (right-hand side). The labels (a) throughout (d) correspond with those in Figs. 6 and 8 ($\alpha = \mathbf{Q} \cdot \mathbf{r}$).

of the SDW's is concerned, the lower temperature has caused its change for Cr 0.6 at. % V and Cr 2.5 at. % V, while the shape of the SDW for the Cr 5.3 at. % V sample has remained unchanged, see Fig. 10. This kind of change for the least- and the medium-concentrated samples indicates the regions that at 295 K were paramagnetic had become ordered (with the SDW structure) at 4.2 K in accord with the phase diagram.² Such behavior has not been, however, found for the most-concentrated sample. This, in our opinion, supports our supposition that the residual SDW's observed in this sample at 295 K have their origin in the internal strain as well as in the interaction with grain boundaries (the latter also enhance SDW's).

Finally, a comparison of the results obtained from the spectra registered for single-crystal and polycrystalline samples at 4.2 K enables the following observations:

- (a) H_{\max} values of the polycrystalline samples are enhanced in comparison with those found for the single crystals.
- (b) The enhancement may be partly due to the internal strain and partly to grain boundaries, both present in our polycrystalline samples.

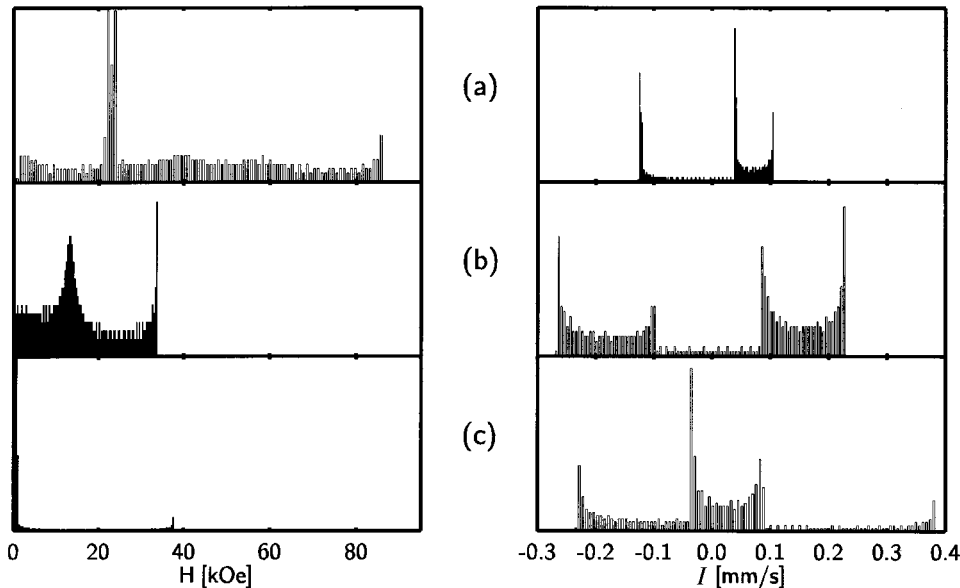


FIG. 9. The histograms of the hf field (left-hand side) and of the isomer shift (right-hand side) distributions derived from the spectra shown in Fig. 8.

(c) $\langle H \rangle$ values of the polycrystalline samples are smaller than the corresponding ones found for the single crystals.

(d) Higher-order harmonics of SDW's and CDW's have enhanced values for polycrystalline samples as compared with their counterparts for the single crystals.

V. CONCLUSIONS

The results obtained in the present study, have shown that ^{119}Sn Mössbauer spectroscopy, which was used here to investigate single crystals of CrV alloys, is a suitable tool in the investigation of SDW's and CDW's in this material. Based on the results the following information has been revealed.

(1) In single-crystal chromium vanadium, doping results in a linear decrease of the maximum hf field (the amplitude of the SDW) as well as that of the average hf field.

(2) The decrease of the normalized values of these two quantities is within 10% the same as that of the Néel temperature, average magnetic moment, and the incommensurability wave vector, and is equal to 0.3 per at. % V.

(3) The spectral parameters H_{max} and $\langle H \rangle$ have been shown to be directly related to the density of states at the Fermi surface.

Conclusions (1) and (2) support the canonical model for the SDW antiferromagnetism in chromium.

ACKNOWLEDGMENTS

One of us (S.M.D.) thanks the Alexander von Humboldt-Stiftung, Bonn and the International Atomic Energy Agency, Vienna for financial support within which the low-temperature measurements described in this paper were carried out.

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