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Spin-density waves in Cr under a magnetic field

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Abstract. The twisting of the spin-density wave (SDW) in Cr in the transverse SDW phase has been examined by neutron scattering experiments in a magnetic field. Twisting beyond experimental errors was observed, but the twisting angle is very small compared to the rotation angle of the SDW in the magnetic field. This result is consistent with the fact that the transverse spin fluctuation in the Cr SDW is strongly suppressed at low excitation energies. The anisotropy energy of the SDW in Cr is also discussed.

1. Introduction

In the transverse spin-density-wave (T-SDW) phase in Cr (121 K $< T <$ 311.5 K), the amplitude of the magnetic moment is sinusoidally modulated along the SDW wave vector $\boldsymbol{Q}$ which is parallel to a cubic axis and the direction of spin polarization is parallel to another cubic axis. Werner *et al*, however, found that the polarization direction of the SDW rotates from the cubic axis when a magnetic field is applied parallel to the spin-polarization direction [1, 2] because the susceptibility difference $\Delta\chi = \chi_\perp - \chi_\parallel$ is positive. To explain the experimental data, they proposed two models of the SDW under a magnetic field. One is based on the thermal activation of the spin orientation in small domains. Another model assumes the existence of the pinning sites to fix the orientation of the spin at particular points in the crystal.

Our interest in the transverse SDW in Cr is in the formation of a twisted SDW under the influence of a magnetic field. The SDW in Cr has rather large magnetic moments in the loop of the SDW and far smaller moments around the node. An anisotropy energy and a magnetic field would act differently in the loop and at the node of the SDW, and the angle of rotation due to the applied magnetic field would depend on the position of the SDW. On the other hand, recent neutron inelastic scattering measurements revealed various interesting features of the magnetic excitations in Cr [3–8]. One peculiar feature is that the longitudinal spin fluctuation (fluctuations of the SDW amplitude) is predominant in the low-frequency excitation—i.e., the transverse spin fluctuation, which is the only magnetic excitation in a localized spin system, is difficult to excite [9, 10]. Since the twisting of the SDW can be considered to be a static transverse spin modulation of the SDW, it is interesting to study how easy (or hard) it is to form the twisted SDW.

2. Observation of twisting in a SDW via neutron scattering

In this section, we explain how to examine the twisting of the SDW via a neutron scattering method. Let us take the xy-plane as the scattering plane and the magnetic field as being applied along the z-axis. We consider here the specimen with a single-$\boldsymbol{Q}$ magnetic domain

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and with $\boldsymbol{Q}$ parallel to the x-direction. (Our experiment was carried out using a single-$\boldsymbol{Q}$-domain sample.) There still exist two $\boldsymbol{S}$-domains in the T-SDW phase, domains in which the spin-polarization direction is parallel to the y-axis (the $\boldsymbol{S}_y$-domain) and the z-axis ($\boldsymbol{S}_z$-domain), respectively. The spins with the polarization direction parallel to the z-axis rotate when the magnetic field is applied along the z-axis. In neutron scattering experiments, only the spin component perpendicular to the scattering vector contributes to the magnetic scattering intensity. Near the 0 1 0 reciprocal-lattice point, since the scattering vector is parallel to the y-axis, we can observe only the z-component of the spins in the $\boldsymbol{S}_z$-domain. (The spins of the $\boldsymbol{S}_y$-domain are parallel to the scattering vector; thus one observes no magnetic scattering from the $\boldsymbol{S}_y$-domain around 0 1 0.) If we write the averaged rotation angle as θ_0, the magnetic satellite intensity $I(\theta_0)$ at $\pm\delta\,1\,0$ is proportional to $I_0\cos^2\theta_0$, where I_0 is the intensity without any magnetic field. Thus, as the rotation angle increases, the satellite peak intensities at $\pm\delta\,1\,0$ decrease. Note that the satellite peak intensities at $1\pm\delta\,0\,0$ are independent of the rotation angle because the spin-polarization vectors in both $\boldsymbol{S}$-domains are always perpendicular to the scattering vector, which is parallel to the x-direction around the 1 0 0 reciprocal-lattice point.

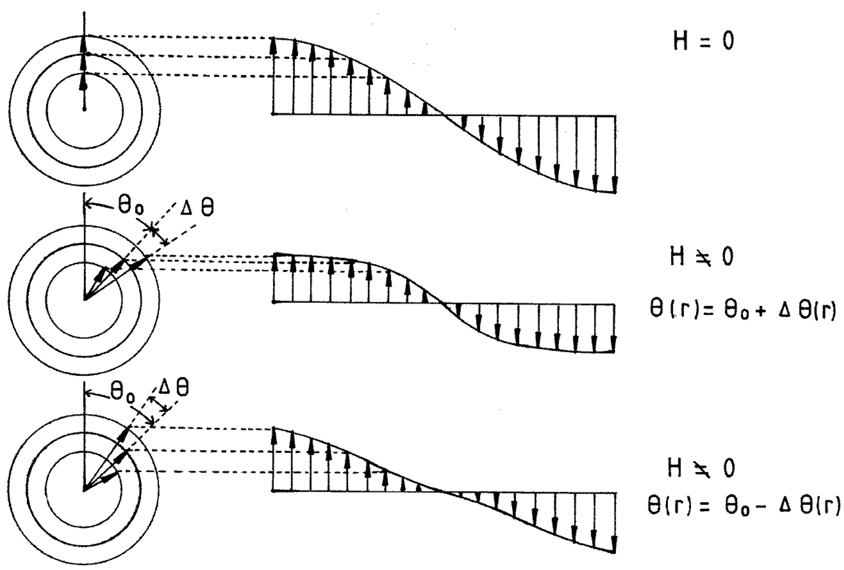

Figure 1. Rotation angles of the SDW moments and the spin component perpendicular to the scattering vector. The third-harmonic component mainly results from the shape change of the SDW due to the twisting.

Now let us consider the twisting of the SDW. If twisting happens, the rotation angle depends on the atomic position $\boldsymbol{r}$ and is written as $\theta(\boldsymbol{r}) = \theta_0 \pm \Delta\theta(\boldsymbol{r})$. Since we are considering the difference between the rotation angle in the loop and at the node of the SDW, the main term of $\Delta\theta(\boldsymbol{r})$ has a period that is twice that of the primary SDW and is written as $\Delta\theta(\boldsymbol{r}) = \sigma\cos 2\boldsymbol{\delta}\cdot\boldsymbol{r}$, where σ is the amplitude of the twisting angle and $\delta = \pi/a - Q$ where a is the lattice parameter. The positive (negative) sign indicates that the rotation angle is the maximum (minimum) in the loop of the SDW. The z-component of the spins can then be described as

$$\boldsymbol{S}_z(\boldsymbol{r}) = \boldsymbol{S}_0\cos(\theta_0 \pm \Delta\theta(\boldsymbol{r}))\ \cos\boldsymbol{Q}\cdot\boldsymbol{r}$$

where $\boldsymbol{S}_0$ is the amplitude of the SDW. After a simple calculation and neglecting the higher-order terms, we obtain the following expression:

$$\boldsymbol{S}_z(\boldsymbol{r}) = \boldsymbol{S}_0(J_0(\sigma)\cos\theta_0 \mp J_1(\sigma)\sin\theta_0)\cos\boldsymbol{Q}\cdot\boldsymbol{r} \mp \boldsymbol{S}_0 J_1(\sigma)\sin\theta_0\cos 3\boldsymbol{Q}\cdot\boldsymbol{r}$$

where J_n indicates the nth-order Bessel function. (See the appendix for the derivation of this equation.) Thus, the twisting of the SDW is mainly observed as a third-harmonic component of the SDW. This situation is schematically illustrated in figure 1.

However, pure Cr already has a third-harmonic component of the SDW (amplitude Δ_3) without there being any magnetic field applied and the magnetic moment is written as [6]

$$\boldsymbol{S}(\boldsymbol{r}) = \boldsymbol{S}_0\cos\boldsymbol{Q}\cdot\boldsymbol{r} - \Delta_3\cos 3\boldsymbol{Q}\cdot\boldsymbol{r}$$

where the minus sign in the second term comes from the observed phase relation between the primary wave and the third-harmonic component of the Cr SDW. Therefore the twisting of the SDW is examined through the change of the third-harmonic satellite intensities relative to the primary satellite intensities as

$$\frac{I(3\delta)}{I(\delta)} = \frac{(\Delta_3\cos\theta_0 \pm S_0 J_1(\sigma)\sin\theta_0)^2}{S_0^2 J_0(\sigma)^2\cos^2\theta_0} = \frac{\Delta_3^2}{S_0^2 J_0(\sigma)^2}\left(1 \pm \frac{S_0 J_1(\sigma)}{\Delta_3}\tan\theta_0\right)^2. \tag{1}$$

The second term includes a strong enhancement factor S_0/Δ_3 and the value of the third-harmonic component relative to the primary wave is very sensitive to the twisting angle σ.

Note that there is another contribution to the third-order satellite intensity arising from the cross term of the primary SDW and the strain wave (the second-harmonic CDW in Cr). This term is observed at the $1 \pm 3\delta$, 0, 0 satellite positions, but the contribution from the cross term is zero at the $\pm 3\delta$, 1, 0 satellite positions, where we study the twisting of the SDW; the scattering cross section for the strain wave is proportional to the term $(\boldsymbol{K}\cdot\boldsymbol{\Delta}_2)^2$, where $\boldsymbol{K}$ is a scattering vector and Δ_2 indicates the amplitude of the strain wave. Since the strain wave is a longitudinal wave, Δ_2 is parallel to the $\boldsymbol{Q}$-vector of the SDW. Thus the term $(\boldsymbol{K}\cdot\boldsymbol{\Delta}_2)^2$ is zero around $0, 1, 0$ in the present experimental conditions.

3. Measurements

Actual measurements of the twisting of the SDW were carried out using a sample of very high quality grown from the vapour in the reduction of chromium iodide. The sample was field cooled through T_N under a magnetic field of 4 T applied along the [1 0 0] axis of the Cr single crystal to grow the single-$\boldsymbol{Q}$-domain sample. Under these conditions, the $\boldsymbol{Q}$-axis of the SDW of the dominant domain is parallel to the [1 0 0] axis. The volume fraction of this domain was about 99% of the total volume as determined from the satellite peak intensity distribution. Then the sample was set so that the [0 0 1] axis was perpendicular to the scattering plane and the magnetic field was applied to the direction parallel to the [0 0 1] axis to study the twisting of the SDW.

Integrated intensities of the primary satellite and the third-harmonic component are plotted in figure 2 as functions of the square of the magnetic field. Integrated intensities were determined via the best-fitting curve of the satellite peaks. In this fitting, a Gaussian curve was used with a fixed peak position and a fixed line width. The intensity of the primary SDW component decreases linearly with the square of the magnetic field for $H^2 \leqslant 1$ T^2. This is consistent with previous work [1, 2]. The third-harmonic component decreases faster than the primary SDW component beyond the statistical errors, indicating that the minus sign in equation (1) is correct, i.e., the rotation angle around the node is larger than that in the loop of the SDW. To estimate the twisting angle σ quantitatively, we used the reported

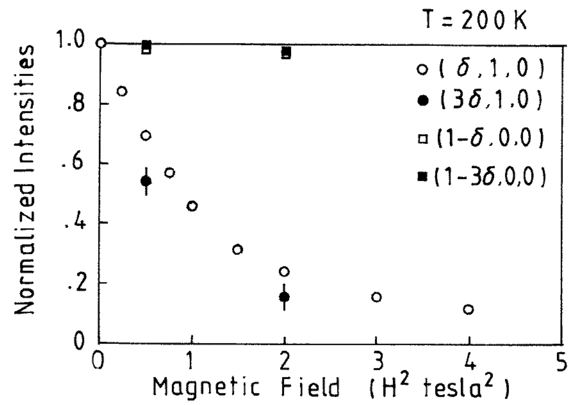

Figure 2. The field dependence of integrated intensities. ○: the primary satellite studied at $(-\delta\,1\,0)$; ●: the third harmonics at $(-3\delta\,1\,0)$; □: the primary satellite at $(1-\delta\,0\,0)$; ■: the third harmonics at $(1-3\delta\,0\,0)$.

value $\Delta_3/S_0 = 1.65 \times 10^{-2}$ [11, 12] and $\theta_0 = 33^\circ$ which is estimated from the observed decreasing of the primary SDW intensity at $H = 0.71$ T. Then we obtain $\sigma = 0.3^\circ$ ($\pm 0.1^\circ$) at $H = 0.71$ T. The same calculation for $H = 1.41$ T gives $\theta_0 = 60.3^\circ$ and $\sigma = 0.2^\circ$ ($\pm 0.1^\circ$). These values appear to be very small compared to the rather remarkable effect. This is due to the factor $S_0/\Delta_3 \sim 61$. For example, a twisting angle of about 3° is enough to cancel the third-harmonic component of Cr completely at $H = 0.71$ T. Thus the twisting of the SDW is very difficult to produce, as is expected from the magnetic excitation measurements as mentioned above.

On the basis of the present experimental data, we can discuss some properties of an anisotropy energy in the Cr SDW. The primary satellite peak intensity decreases to less than half of the original value, indicating that the rotation angle of the magnetic moment is larger than $\pi/4$. In the cubic system, each cubic axis should be an easy axis, if one of the cubic axes is the easy axis and the anisotropy energy has a fourfold symmetry. The present results, however, show that this is not the case. The anisotropy energy has a twofold symmetry around the $\boldsymbol{Q}$-vector. The total energy of the SDW under the magnetic field is thus described as

$$E(H) = K_2 \sin^2\theta_0 + K_4 \sin^4\theta_0 + (1/2)\,\Delta\chi H^2 \cos^2\theta_0.$$

The first and second terms are the anisotropy energy and the third indicates the magnetic field energy [13]. As an expression for the anisotropy energy, the first term is not sufficient to explain the rotation of the SDW under the magnetic field because it can be included in the third term (except for a constant energy) if we rewrite the first term using the relation $\sin^2\theta_0 = 1 - \cos^2\theta_0$. The rotation angle would be determined from the condition $\mathrm{d}E/\mathrm{d}\theta_0 = 0$. However, at $H = 0$, θ_0 should be zero. Thus, K_2 must be zero and we obtain the solution

$$\sin^2\theta_0 = \Delta\chi H^2/4K_4.$$

The observed satellite intensity $I(\delta)$ is proportional to $\cos^2\theta_0 = 1 - \sin^2\theta_0 = 1 - \alpha H^2$ from the above equation ($\alpha = \Delta\chi/4K_4$). The H^2-dependence of the decreasing intensity is consistent with the observation at $H^2 < 1$ T^2 as shown in figure 2.

The dominant contribution to the anisotropy energy is considered to be the fourth-order term. The lattice deformation is a good candidate for explaining the fourth-order form of the anisotropy energy. Since a lattice strain usually couples with the square of the magnetic moment, the elastic energy is proportional to the fourth order of the magnetic moment. Furthermore, the local lattice structure of Cr is known to have an orthorhombic symmetry below the Néel temperature [14]. This is also consistent with the twofold symmetry of the anisotropy energy around the $\boldsymbol{Q}$-vector.

Here we treated the anisotropy energy in the same way as for a localized spin system. This may not be suitable in the case of Cr. In the case of an itinerant spin system, we may have to consider the movement of each d electron under the magnetic field. That is beyond the scope of the present paper.

4. The longitudinal magnon in a magnetic field

The magnetic excitation of Cr in the transverse SDW phase was also studied in a magnetic field and the data were compared with those obtained without the magnetic field. Under a high-strength magnetic field, polarization of the magnetic moments is nearly parallel to the scattering vector around the 0 1 0 reciprocal point. The magnetic excitation observable under these conditions is fully ascribable to the transverse spin fluctuation. However, without the magnetic field, the longitudinal spin fluctuation contributes to a part of the inelastic scattering intensity. (If we assume the equal distribution of the $\boldsymbol{S}$-domains, a quarter of the intensity is ascribable to the longitudinal spin fluctuation.) Thus, we can estimate the contribution of the longitudinal spin-fluctuation component by comparing the inelastic scattering intensities with and without the magnetic field. In figure 3, experimental data obtained with constant-E scans around the $\pm\delta$ 1 0 satellite positions are given for different energy transfers. For the energy transfer of 2 meV, the inelastic scattering intensity without the magnetic field is appreciably stronger than that under the applied field. For 8 meV energy transfer, however, there is no change of the scattering intensity, indicating that although the magnetic excitation at high energy transfer (8 meV) is isotropic, the longitudinal spin fluctuation is predominant for the low-energy excitation. To be sure about this point, we have made the same type of measurement around the $1 \pm \delta$ 0 0 satellite points, where the contributions from the transverse and longitudinal spin fluctuations are not affected by the magnetic field. The observed data are given in figure 4. These features are qualitatively consistent with the work of previous authors [9, 10]. Grier *et al* studied the inelastic scattering intensities around δ 1 0 under the magnetic field of $H = 6$ T [10] and reported that there was no change of the magnon intensity for at $\Delta E = 4$ meV. This energy transfer would be already too high for observing the anisotropy of the magnetic excitations, as was expected from the data of Burke *et al* [9].

5. Conclusions

The twisting of the SDW in the transverse SDW phase in Cr was studied in an applied magnetic field. When the spin-polarization axis rotates to the direction perpendicular to the field direction, the anisotropy energy and the magnetic energy work differently in the loop and at the node of the SDW and twisting of the SDW is expected in the magnetic field.

Experimental data show that the twisting actually exists beyond experimental errors, and the rotation angle around the node is larger than that around the loop, but the twisting angle is very small ($\sim 0.3^\circ$) compared to the averaged rotation angle ($\theta_0 \sim 33^\circ$) in a magnetic field of 0.71 T.

Recent measurements of magnetic excitation in Cr show that the longitudinal spin

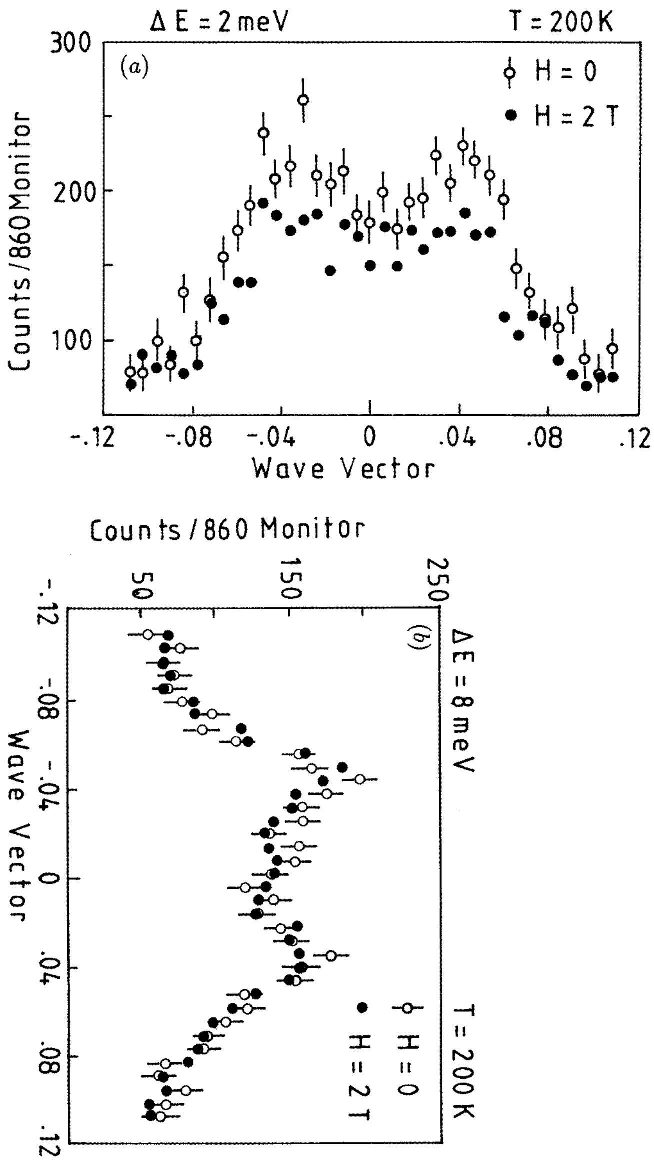

Figure 3. Constant-E measurements of the magnetic excitations obtained at the $(\pm\delta\ 1\ 0)$ satellite peak positions for the energy transfer $\Delta E =$ (a) 2 meV and (b) 8 meV.

fluctuation is predominant and the tranverse spin fluctuation is suppressed at low frequency. We also studied the magnetic excitation of Cr in the T-SDW phase in the magnetic field and confirmed this. Since the twisting of the SDW can be considered to be a static transverse spin fluctuation, the result of the twisting measurement is consistent with the fact that the transverse spin fluctuation is suppressed in the low-frequency magnetic excitation in Cr [9, 10].

The origin of the anisotropy energy of the SDW is also discussed. The lattice deformation associated with the formation of the SDW is a good candidate for explaining

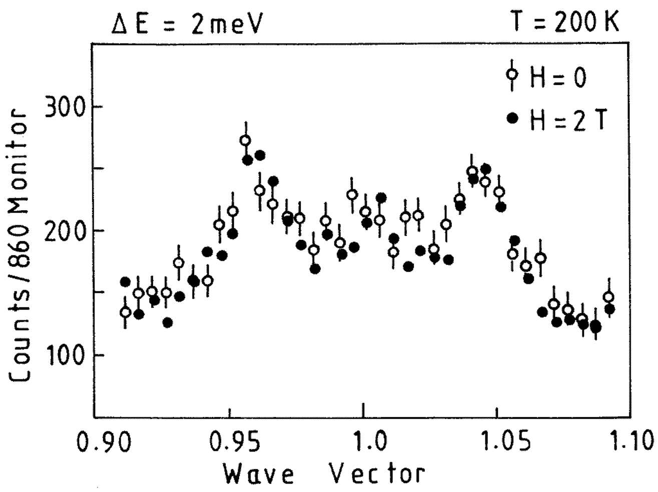

Figure 4. Constant-E measurements of the magnetic excitations obtained at the $(1 \pm \delta\, 0\, 0)$ satellite peak positions for the energy transfer $\Delta E = 2$ meV.

the experimental data.

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Appendix

The z-component of the spins for the $\boldsymbol{S}_z$-domain is written as

$$\boldsymbol{S}_z(\boldsymbol{r}) = \boldsymbol{S}_0 \cos(\theta_0 \mp \Delta\theta(\boldsymbol{r})) \cos \boldsymbol{Q}\cdot\boldsymbol{r} = \boldsymbol{S}_0 \cos \boldsymbol{Q}\cdot\boldsymbol{r}\{\cos\theta_0 \cos\Delta\theta(\boldsymbol{r}) \mp \sin\theta_0 \sin\Delta\theta(\boldsymbol{r})\}$$

where

$$\Delta\theta(\boldsymbol{r}) = \sigma \cos 2\boldsymbol{\delta}\cdot\boldsymbol{r}.$$

The terms $\cos\Delta\theta(\boldsymbol{r})$ and $\sin\Delta\theta(\boldsymbol{r})$ are rewritten using the equalities:

$$\cos(z\cos\zeta) = J_0(z) + 2\sum_n (-1)^n J_{2n}(z)\cos(2n\zeta)$$

$$\sin(z\cos\zeta) = 2\sum_n (-1)^n J_{2n+1}(z)\cos((2n+1)\zeta).$$

Since the amplitude σ is considered to be small compared to the rotation angle θ_0, we can neglect the higher-order terms J_n ($n \geqslant 2$). Then we obtain

$$\begin{aligned}\boldsymbol{S}_z(\boldsymbol{r}) &= \boldsymbol{S}_0 \cos\boldsymbol{Q}\cdot\boldsymbol{r}\{J_0(\sigma)\cos\theta_0 \mp 2J_1(\sigma)\sin\theta_0\cos 2\boldsymbol{\delta}\cdot\boldsymbol{r}\}\\ &= \boldsymbol{S}_0(J_0(\sigma)\cos\theta_0 \mp J_1(\sigma)\sin\theta_0)\cos\boldsymbol{Q}\cdot\boldsymbol{r} \mp \boldsymbol{S}_0 J_1(\sigma)\sin\theta_0\cos 3\boldsymbol{Q}\cdot\boldsymbol{r}\end{aligned}$$

where we have used the following equalities:

$$\cos 2\boldsymbol{\delta}\cdot\boldsymbol{r} = \cos 2\boldsymbol{Q}\cdot\boldsymbol{r} \qquad 2\cos\boldsymbol{Q}\cdot\boldsymbol{r}\cos 2\boldsymbol{Q}\cdot\boldsymbol{r} = \cos\boldsymbol{Q}\cdot\boldsymbol{r} + \cos 3\boldsymbol{Q}\cdot\boldsymbol{r}.$$

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