

A detailed analysis of dipolar interactions in arrays of bi-stable magnetic nanowires

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Abstract

The investigation of the role of interactions in magnetic wire arrays is complex and often involves substantial simplifications. In this paper analytical expressions taking into consideration the geometry of the wires and dipolar interactions between them have been obtained. An expansion of these terms, at first order, can be easily evaluated and shows a good agreement with the total expression for the energy. The extent of the interwire magnetostatic coupling has also been investigated, and it is shown that the number of wires required to reach a size independent magnetic state in the array strongly depends on the relative magnetic orientation of the wires.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

During the last decade, regular arrays of magnetic nanoparticles have been deeply investigated. Besides the basic scientific interest in the magnetic properties of these systems, there is evidence that they might be used in the production of new magnetic devices [1, 2]. Different geometries have been considered, including dots, rings, tubes and wires. Recent studies on such structures have been carried out with the aim of determining the stable magnetized state as a function of the geometry of the particles [3–5]. In particular, the study of highly ordered arrays of magnetic wires with diameters typically in the range of tens to hundreds of nanometers is a topic of growing interest [6–9]. This is a consequence of the development of experimental techniques that lead to fabrication in a controllable and ordered way of such arrays [10, 11]. The high ordering of the array, together with the magnetic nature of nanowires, gives rise to outstanding cooperative properties of fundamental and technological interest [12].

Bi-stable nanowires are characterized by square-shaped hysteresis loops defined by the abrupt reversal of the

magnetization between two stable remanent states [13, 14]. In such systems, effects of interparticle interactions are in general complicated by the fact that the dipolar fields depend upon the magnetization state of each element, which in turn depends upon the fields due to adjacent elements. Therefore, the modeling of interacting arrays of nanowires is often subject to strong simplifications, for example, modelling the wire using a one-dimensional modified classical Ising model [14, 15]. Zhan *et al* [16] used the dipole approximation including additionally a length correction. Velázquez and Vázquez [17, 18] considered each microwire as a dipole, in such a way that the axial field generated by a microwire is proportional to its magnetization. Nevertheless, this model is merely phenomenological since the comparison of experimental results with a strictly dipolar model shows that the interaction in the actual case is more intense. They have also calculated the magnetostatic field and expanded it in multipolar terms [19], showing that the non-dipolar contributions of the field are non-negligible for distances considered in experiments. Recently, the energy of magnetostatic interaction between two magnetic elements of arbitrary shape was derived within the framework

of a Fourier space approach by Beleggia *et al* [20, 21]. In spite of the extended study of the dipolar interactions, a detailed calculation of these interactions in arrays of magnetic structures has not been presented yet. Also micromagnetic calculations [22, 23] and Monte Carlo simulations [24] have been developed. However, these two methods permit us to investigate arrays with just a few wires.

The purpose of this work is to investigate the magnetic behavior of arrays of bi-stable ferromagnetic nanowires as a function of the relative magnetic orientation of the interacting wires, and the geometrical parameters involved. We start by developing an analytical model for arrays which includes shape anisotropy (or dipolar self-energy) of each wire as well as the full long-range magnetostatic interaction within the array. Our model goes beyond the dipole–dipole approximation and leads us to obtain an analytical expression for the interaction in which the lengths and radii of the wires are taken into account. Using those expressions we developed Monte Carlo simulations to investigate the hysteretic behavior of the array.

2. Continuous magnetization model

Geometrically, nanowires are characterized by their radius, R , and length, L . The description of an array of N wires based on the investigation of the behavior of individual magnetic moments becomes numerically prohibitive. In order to circumvent this problem we use a continuous approach and adopt a simplified description in which the discrete distribution of magnetic moments in each wire is replaced with a continuous one, defined by a function $\mathbf{M}(\mathbf{r})$ such that $\mathbf{M}(\mathbf{r})\delta V$ gives the total magnetic moment within the element of volume δV centered at \mathbf{r} . We recall that E_{tot} is generally given by the sum of three terms corresponding to the magnetostatic, E_{dip} , the exchange, E_{ex} , and the anisotropy contributions. Here we are interested in soft or polycrystalline magnetic materials, in which case the anisotropy is usually disregarded [7].

The total magnetization can be written as $\mathbf{M}(\mathbf{r}) = \sum_{i=1}^N \mathbf{M}_i(\mathbf{r})$, where $\mathbf{M}_i(\mathbf{r})$ is the magnetization of the i th nanowire. In this case, the magnetostatic potential $U(\mathbf{r})$ splits up into N components, $U_i(\mathbf{r})$, associated with the magnetization of each individual nanowire. Then, the total dipolar energy can be written as $E_{\text{dip}} = \sum_{i=1}^N E_{\text{dip}}(i) + \sum_{i=1}^{N-1} \sum_{j=i+1}^N E_{\text{int}}(i, j)$, where

$$E_{\text{dip}}(i) = \frac{\mu_0}{2} \int \mathbf{M}_i(\mathbf{r}) \cdot \nabla U_i(\mathbf{r}) \, dv \quad (1)$$

is the dipolar contribution to the self-energy of nanowire i th, and

$$E_{\text{int}}(i, j) = \mu_0 \int \mathbf{M}_i(\mathbf{r}) \nabla U_j(\mathbf{r}) \, dv \quad (2)$$

is the dipolar interaction between nanowires i and j . In the dipolar contribution to the self-energy an additive term independent of the configuration has been left out [25].

In this work we investigate bi-stable nanowires in which case [25], $E_{\text{ex}} = \sum_{i=1}^N E_{\text{ex}}(i) = 0$. On the basis of this assumption, the total energy of the array can be written as

$$E_{\text{tot}} = \sum_{i=1}^N E_{\text{self}}(i) + \sum_{i=1}^{N-1} \sum_{j=i+1}^N E_{\text{int}}(i, j), \quad (3)$$

where $E_{\text{self}}(i) = E_{\text{dip}}(i)$ is the dipolar self-energy of each wire, and $E_{\text{int}}(i, j)$ is the dipolar interaction energy between wires i th and j th.

2.1. Total energy calculation

We now proceed to the calculation of the energy terms in the expression for E_{tot} . Results are given in units of $\mu_0 M_0^2 V$, i.e. $\tilde{E} = E/\mu_0 M_0^2 V$, where $V = \pi R^2 L$ is the volume and M_0 is the saturation magnetization of each nanowire.

In order to evaluate the total energy, it is necessary to specify the functional form of the magnetization for each nanowire. We consider wires with an axial magnetization defined by $\mathbf{M}_i(\mathbf{r}) = M_0 \sigma_i \hat{\mathbf{z}}$, where $\hat{\mathbf{z}}$ is the unit vector parallel to the axis of the nanowire and σ_i takes the values ± 1 , allowing the wire i to point up ($\sigma_i = +1$) or down ($\sigma_i = -1$) along $\hat{\mathbf{z}}$.

2.1.1. Self-energy of a nanowire. The reduced dipolar self-energy has been calculated by Tandon *et al* [26] and takes the form

$$\tilde{E}_{\text{self}}(i) = \frac{1}{2} \left(1 + \frac{8R}{3\pi L} - F_{21} \left[-\frac{4R^2}{L^2} \right] \right), \quad (4)$$

where $F_{21}[x] = F_{21}[-1/2, 1/2, 2, x]$ is a hypergeometric function. Note that in equation (4) the energy of each wire depends only on the ratio R/L . As an example, when we consider a Ni wire with $L = 1 \mu\text{m}$, $R = 20 \text{ nm}$ and $L/R = 50$, the self-energy is $E_{\text{self}} = 19.45 \text{ eV}$.

2.1.2. Interwire magnetostatic coupling. The interaction between two nanowires is obtained using the magnetostatic field experienced by one of the wires due to the other so that the final result reads

$$\tilde{E}_{\text{int}}(i, j) = 2\sigma_i \sigma_j \int_0^\infty \frac{dq}{q^2} J_0 \left(\frac{q S_{ij}}{L} \right) J_1^2 \left(\frac{qR}{L} \right) (1 - e^{-q}), \quad (5)$$

where J_p is a Bessel function of the first kind and p order and S_{ij} is the center-to-center distance between the magnetic nanowires i and j . The previous equation allows us to write the interaction energy of two wires as $\tilde{E}_{\text{int}}(i, j) = \sigma_i \sigma_j \tilde{E}_{\text{int}}(S_{ij}) = \pm \tilde{E}_{\text{int}}(S_{ij})$, where the sign $+$ ($-$) corresponds to $\sigma_i = \sigma_j$ ($\sigma_i \neq \sigma_j$).

Equation (5) has been previously obtained by Beleggia *et al* (see equation (59) in [20]) considering a more general approach based on Fourier transforms of the magnetization.

2.2. Results

2.2.1. Two-wire system. The general expression giving the interaction between wires with axial magnetization is given by equation (5). This expression has to be solved numerically. However, wires that motivate this work [6–9] satisfy $L/R \gg 1$, leading us to expand J_1 as

$$J_1(x) = \frac{x}{2} - \frac{x^3}{16} + \sum_{k=2}^{\infty} \frac{(-1)^k (x/2)^{1+2k}}{k! \Gamma(k+2)}. \quad (6)$$

Then we can approximate equation (5) by

$$\tilde{E}_{\text{int}}^\lambda(S_{ij}) = \frac{R^2}{2LS_{ij}} \sum_{\lambda=1}^{\infty} g_\lambda, \quad (7)$$

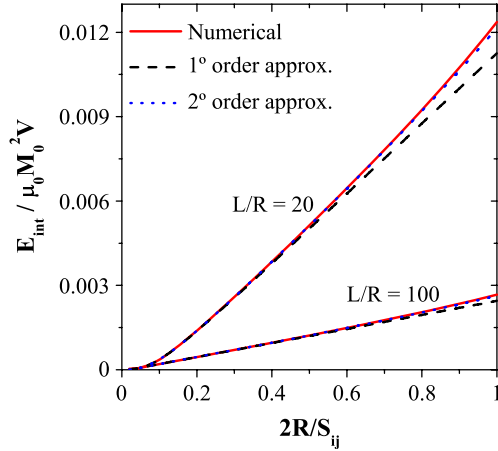


Figure 1. Energy of interaction between two identical nanowires with parallel axial magnetization. The solid line (red online) corresponds to the numerical integration of equation (5), the dashed line (black) corresponds to the first-order approximation of equation (7) and the dotted line (blue online) corresponds to the second-order approximation of equation (7).

where λ indicates the order of the expansion. As an example, the first and second terms in the sum are

$$g_1 = 1 - \frac{1}{\alpha_1} \quad (8)$$

and

$$g_2 = \frac{R^2}{4S_{ij}^2} \left(1 - \frac{\alpha_2}{\alpha_1}\right) + \frac{9R^4}{64S_{ij}^4} \left(1 - \frac{\alpha_3}{\alpha_1}\right), \quad (9)$$

where $\alpha_1 \equiv \sqrt{1 + L^2/S_{ij}^2}$, $\alpha_2 \equiv (1 - 2L^2/S_{ij}^2)/\alpha_1^4$ and $\alpha_3 \equiv (1 + 8L^4/3S_{ij}^4 - 8L^2/S_{ij}^2)/\alpha_1^8$. Figure 1 illustrates the energy of interaction between two identical nanowires with parallel axial magnetization as a function of $2R/S_{ij}$. When the two wires are in contact, $2R/S_{ij} = 1$; when they are infinitely separated, $2R/S_{ij} = 0$. In this figure the solid line represents the numerical integration of the interaction energy, equation (5), the dashed line is given by the first-order approximation of this energy, equation (7), and the dotted line corresponds to the second-order approximation. From this figure we can conclude that the first term in the expansion in equation (7) gives a very good approach to equation (5) for $2R/S_{ij} \leq 0.6$, and $L/R \gg 1$. As an illustration, when we consider two Ni wires with $L = 1 \mu\text{m}$, $R = 20 \text{ nm}$ and $S_{ij} = 100 \text{ nm}$, we obtain $E_{\text{int}}(S_{ij}) = 4.22 \text{ eV}$ and $E_{\text{int}}^1(S_{ij}) = 4.18 \text{ eV}$.

In order to quantify the relative importance of the interaction energy we calculated the ratio between the self-energy and the magnetostatic interaction energy between two identical nanowires,

$$\eta = \frac{\tilde{E}_{\text{int}}(S_{ij})}{2\tilde{E}_{\text{self}}} \quad (10)$$

Figure 2 defines the geometry of the two-wire system for which $\eta = 0.2, 0.1, 0.01$ and 0.001 . From this figure we observe a strong dependence of the interaction energy on the geometry of the two-wire system. As an illustration, when we consider two nanowires with $L = 1 \mu\text{m}$, $R = 20 \text{ nm}$ and

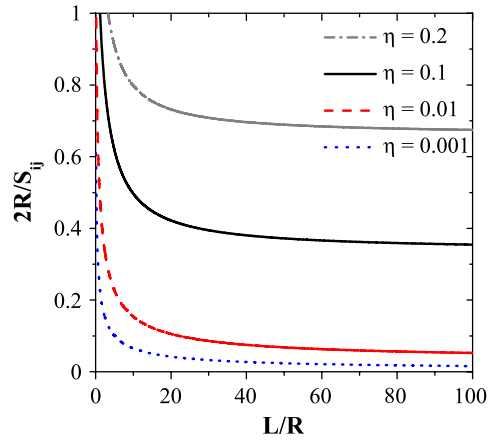


Figure 2. η values as a function of the geometry of the two-wire system.

$L/R = 50$, if we look at an almost non-interacting regime, $\eta = 0.01$, $2R/S_{ij} = 0.068$, and then the two wires have to be at least 590 nm apart. For this geometry the interaction energy is about 1% of the self-energy. However, for the same L and R , if the wires are 58 nm apart ($2R/S_{ij} = 0.69$), the interaction energy is about 20% of the self-energy ($\eta = 0.2$).

2.2.2. Wire array. The focus of this work is on clarifying the role of dipolar interactions in the magnetic behavior of a square array of nanowires. With the previous relations for a two-wire system, we are now in a position to investigate the effect of the interwire magnetostatic coupling in the array. Calculations for the total interaction energy $\tilde{E}_{\text{array}}^{\pm}(N)$ of the $N = n \times n$ square array are shown in the appendix, and lead us to write

$$\begin{aligned} \tilde{E}_{\text{array}}^{\pm}(N) = & 2n \sum_{p=1}^{n-1} (n-p)(\pm 1)^p \tilde{E}_{\text{int}}(pd) \\ & + 2 \sum_{p=1}^{n-1} \sum_{q=1}^{n-1} (n-p)(n-q)(\pm 1)^{p+q} \tilde{E}_{\text{int}}(d\sqrt{p^2+q^2}), \end{aligned}$$

where $+$ ($-$) refers to ferromagnetic (antiferromagnetic) magnetic ordering of the nanowires in an array with nearest-neighbor distance d , and \tilde{E}_{int} is the energy of interaction between two identical magnetic elements of arbitrary shape. In particular, for magnetic nanowires \tilde{E}_{int} is given by equation (5). Note that in an array S_{ij} is a function of d . In the antiferromagnetic configuration the magnetizations of nearest-neighbor nanowires point in opposite directions defining the lowest energy state of the array [27]. Figure 3 illustrates the behavior of $W_{\text{array}}^{\pm}(N) = \tilde{E}_{\text{array}}^{\pm}(N)/N$ as a function of N in a ferromagnetic (solid line) and an antiferromagnetic (dashed line) array. We consider an array of identical wires with $R = 20 \text{ nm}$ and $L = 1 \mu\text{m}$ and $d = 50 \text{ nm}$. We can see that a large number of wires ($N \approx 10^6$), corresponding to a sample of $\approx 0.01 \text{ mm}^2$, is required for reaching convergence of $W_{\text{array}}^+(N)$. However, in view of cancellations originating in the different signs of the parallel and antiparallel interactions, the antiferromagnetic configuration converges faster, requiring only of the order of 10^2 wires and a sample of $\approx 1 \mu\text{m}^2$.

We also investigate, for the same wires ($R = 20 \text{ nm}$ and $L = 1 \mu\text{m}$), the variation of the asymptotic value of

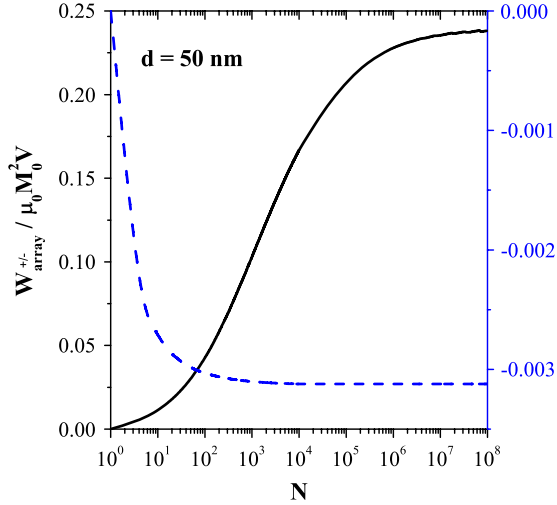


Figure 3. $W_{\text{array}}^{\pm}(N)$ (solid (black), ferromagnetic array; dashed (blue), antiferromagnetic array) in a square array of identical wires ($R = 20$ nm and $L = 1$ μm). We consider different numbers of wires in the array and $d = 50$ nm. The scale for the antiferromagnetic array is on the right.

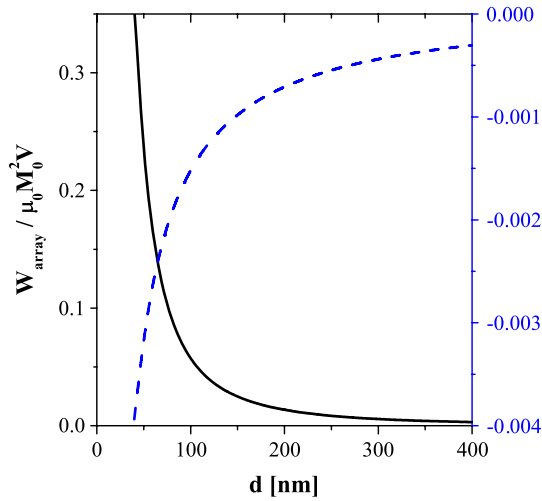


Figure 4. Asymptotic values of the interaction energy in a square array obtained with $N = 10^6$ (solid line, ferromagnetic array) and $N = 10^2$ (dashed line, antiferromagnetic array).

$W_{\text{array}}^{\pm}(N)$ as a function of the nearest-neighbor distance, d , in ferromagnetic (solid line) and antiferromagnetic (dashed line) arrays. Figure 4 illustrates our results showing that in the ferromagnetic array, interaction effects decay but extend over long distances. Figures 3 and 4 agree with conclusions from experiments by Nielsch *et al* [6] who assume that, due to the high aspect ratio of the magnetic nanowires in a hexagonal array, the stray field interaction extends over several nearest-neighbor distances.

3. Monte Carlo simulations

As a consequence of the large aspect ratio of the wires investigated, the anisotropy that they present is mainly shape anisotropy. In this case, the individual wires can be considered

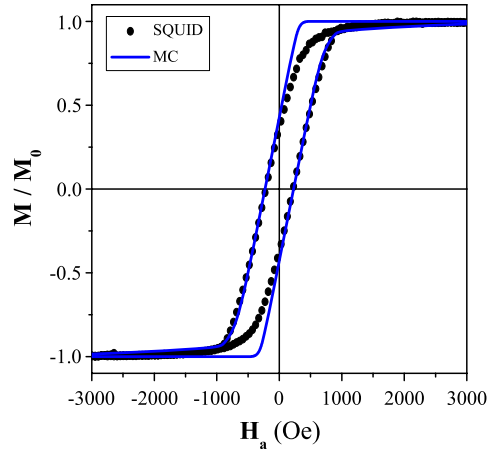


Figure 5. Hysteresis loops as measured with a SQUID (black dots) and obtained from numerical simulations (solid/blue line) with the external field applied parallel to the wire direction.

as nearly single-domain structures with two stable states: the magnetic moment pointing up or down. However, the behavior of the array as a whole differs from a pure bi-stable magnetic state due to the magnetostatic interactions between the nanowires. In order to model the hysteresis loop of the array we develop Monte Carlo (MC) simulations considering the expression for the magnetostatic interaction among wires obtained in section 2. Therefore the internal energy of a hexagonal array with N identical wires can be written as

$$E = \mu_0 M_0 V \left(\sum_{i=1}^{N-1} \sum_{j=i+1}^N M_0 \tilde{E}_{\text{int}}^1(S_{ij}) - (H_a + H_c) \sum_{i=1}^N \sigma_i \right).$$

The first term in the above equation is the magnetostatic interaction of all pairs of magnetic wires. The coupling $\tilde{E}_{\text{int}}^1(S_{ij})$ is given by equation (7). The second term corresponds to the contribution of an external magnetic field, H_a , applied along the axis of the wire and the third term, H_c , represents the magnetic shape anisotropy of a single wire, giving its coercivity. Note that the information about the geometrical arrangement of wires within the array is given by relative distances S_{ij} . Besides, coercivity and saturation magnetization values have to be settled for calculations. In our simulations we used $M_0 = 480$ emu cm^{-3} , a standard value for Niquel, and $H_c = 215$ Oe, obtained from experimental results shown in figure 5 in this paper. The hysteresis loops were simulated with the external field in the direction of the wire axis. The initial state, at $H_a = 4.0$ kOe, higher than the saturation field, has a configuration in which all the magnetic moments were aligned with the external field. The field was then linearly decreased at a rate of 300 Monte Carlo steps for $\Delta H_a = 0.01$ kOe.

Figure 5 shows the hysteresis loop of the Ni nanowire array ($R = 90$ nm, $L = 3.6$ μm and $d = 500$ nm) along the axial direction. The black dots were obtained from measurements [28] with a superconducting quantum interference device (SQUID) and the solid line by MC simulations considering $N = 9699$.

Deviations between SQUID measurements and numerical calculations can originate in the dispersion of lengths and positions of each wire in the array and a reduction in the

homogeneity of the diameter of nanopores [29]. From figure 3 we have obtained that a large number of wires ($N \approx 10^6$) is required for reaching convergence of $W_{\text{array}}^+(N)$. However, with present standard computational capabilities it is not possible to obtain hysteresis loops with N higher than 10000. Thus, if smaller samples are simulated, it is necessary to be careful when comparing with experimental results.

4. Conclusions

By expanding analytical expressions for the magnetostatic interactions between wires, we investigate first-order and second-order approximations to the interaction energies showing the range of validity of these expansions. When the wires are apart at distances much larger than their diameters, the first-order approximation is valid. The energy expressions lead us to investigate the extent of the interwire magnetostatic interactions in a square array. The number of wires required to obtain independent results on the size of the array strongly depends on the relative magnetic ordering of nearest-neighbor wires. For the ferromagnetic array, and due to the additive nature of the dipolar interaction, results strongly depend on the size of the array, and a very significant number of wires, 10^6 , is required to obtain size independent results. Then, the size of the array is an important factor to be consider when different measurements have to be compared. Monte Carlo simulations of hysteresis loops with the corrected magnetostatic interaction among wires give fairly good agreement with experimental measurements.

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Appendix. Total interaction energy of a square array

The total interaction energy of the $N = n \times n$ square array can be written as

$$\tilde{E}_{\text{array}}^{\pm}(n) = \frac{1}{2} \sum_{i=1}^n \sum_{j=1}^n \sum_{k=1}^n \sum_{l=1}^n (\pm 1)^{(k-i)-(l-j)} \times \tilde{E}_{\text{int}} \left(d\sqrt{(k-i)^2 + (l-j)^2} \right),$$

where $+$ ($-$) refers to parallel (antiparallel) nearest-neighbor magnetic orientation of the elements in the array. Note that \tilde{E}_{int} corresponds to the dipolar interaction energy between two identical magnetic elements of arbitrary shape. Here $\tilde{E}_{\text{int}}(0) = 0$, avoiding the self-interaction of the particles. For simplicity we define the following function:

$$f^{\pm}(p, q) = (\pm 1)^{p-q} \tilde{E}_{\text{int}}(d\sqrt{p^2 + q^2}), \quad (\text{A.1})$$

which can be used to write the interaction energy in a compact form; that is

$$\tilde{E}_{\text{array}}^{\pm}(n) = \frac{1}{2} \sum_{i=1}^n \sum_{j=1}^n \sum_{k=1}^n \sum_{l=1}^n f^{\pm}(k-i, l-j). \quad (\text{A.2})$$

We can reduce the number of summations using the following rule:

$$\sum_{i=1}^n \sum_{k=1}^n g(k-i) = ng(0) + \sum_{p=1}^{n-1} (n-p)[g(p) + g(-p)], \quad (\text{A.3})$$

which leads us to write

$$\begin{aligned} \sum_{i=1}^n \sum_{k=1}^n f^{\pm}(k-i, l-j) &= nf^{\pm}(0, l-j) \\ &+ 2 \sum_{p=1}^{n-1} (n-p) f^{\pm}(p, l-j). \end{aligned}$$

Then, the interaction energy, equation (A.2), reduces to

$$\begin{aligned} \tilde{E}_{\text{array}}^{\pm}(n) &= \frac{n}{2} \sum_{j=1}^n \sum_{l=1}^n f^{\pm}(0, l-j) \\ &+ \sum_{p=1}^{n-1} (n-p) \sum_{j=1}^n \sum_{l=1}^n f^{\pm}(p, l-j). \end{aligned} \quad (\text{A.4})$$

Using again the rule (A.3), we can reduce the double sums in equation (A.4), obtaining

$$\begin{aligned} \tilde{E}_{\text{int}}^{\pm}(n) &= \frac{n^2}{2} f^{\pm}(0, 0) + n \sum_{q=1}^{n-1} (n-q) f^{\pm}(0, q) \\ &+ n \sum_{p=1}^{n-1} (n-p) f^{\pm}(p, 0) \\ &+ 2 \sum_{p=1}^{n-1} \sum_{q=1}^{n-1} (n-p)(n-q) f^{\pm}(p, q). \end{aligned}$$

From (A.1) we know that $f^{\pm}(0, 0) \equiv 0$, which leads us to finally obtain

$$\begin{aligned} \tilde{E}_{\text{array}}^{\pm}(n) &= 2n \sum_{p=1}^{n-1} (n-p) (\pm 1)^p \tilde{E}_{\text{int}}(pd) \\ &+ 2 \sum_{p=1}^{n-1} \sum_{q=1}^{n-1} (n-p)(n-q) (\pm 1)^{p-q} \tilde{E}_{\text{int}} \left(d\sqrt{p^2 + q^2} \right). \end{aligned}$$

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