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Accurate determination of second order nonlinear optical coefficients from powder crystal monolayers

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An accurate procedure for the determination of second harmonic generation (SHG) efficiency from powder crystal monolayers is proposed. The method is based on the analysis of the dependence of the SHG output on the powder particle size. Unlike with conventional powder methods, the amount of material required is very small, and the scattering of the primary beam by the powder particles is negligible, allowing for an accurate analysis of the collected data. The experimental results demonstrate that the proposed method can provide reliable values for the nonlinear optical coefficients, particularly in the case of phase-matchable uniaxial materials, for which simple analytical expressions can be derived. © *2011 American Institute of Physics*. [doi:10.1063/1.3592964]

I. INTRODUCTION

Second-order nonlinear optical susceptibilities[1–4] d_{ij} are usually determined by measuring the second-harmonic generation (SHG) efficiencies on single crystals.[5,6] Many times, however, good crystals are not available. In those cases, the powder method developed by Kurtz and Perry[7] is commonly used. The technique gives an estimate of an effective nonlinear coefficient together with an idea of the possibility of attaining the phase matching (PM) condition. The last point requires SHG measurements of various powder samples with different particle sizes.

The Kurtz and Perry method is extremely popular for evaluating or surveying new nonlinear optical materials.[8–17] In fact, it can be verified[18] that the number of citations in the literature has been growing with time during past years (due to the increasing activity in the synthesis of new materials for nonlinear optical applications), and the total amount of citations is close to 2000! However, it is also known that problems can arise with the method when quantitative information about the d_{ij} coefficients is looked for. Care should be taken with regard to the conclusions drawn for the measured materials, because in most cases nothing more than a rough idea of the nonlinear efficiencies can be obtained. In this respect, it is interesting to point out that it is not unusual to find differences of one order of magnitude between the d_{ij} values determined with this method and those found with single crystal techniques.[1,8,19] One fundamental reason for that is that the semiquantitative model of Kurtz and Perry[7] disregards the scattering by the powder particles of the sample. Even when the powder is mixed with a liquid with approximately the same index of refraction as the crystal, the length of the sample is usually long enough to ignore the effect of light scattering. In this article, we show that a slight modification of the Kurtz and Perry technique, together with a careful evaluation of the particle size of the crystalline powder, can yield reliable quantitative data for the d_{ij} coefficients. The method is based on the analysis of the SHG output of a powder crystal monolayer.

SHG by powder crystal monolayers has been scarcely studied.[20–22] The works published on this subject have been essentially experimental[20–22] and have been focused on the analysis of the dependence of the SHG signal on the powder particle size. According to these works,[20,21] in the case of phase-matchable materials, the SHG signal depends linearly on the powder particle size, whereas in non-phase-matchable materials the SHG output is independent of the particle size. This result would allow for a simple classification of a nonlinear optical material during a survey. Nevertheless, no theoretical analysis of the data was carried out in these works. Our first aim was to develop a model that could quantitatively describe those experimental results.

II. THEORETICAL MODEL

We assume that the incoming beam is a plane wave and that the particles are immersed in an index matching liquid[20,21] in order to reduce the scattering as much as possible. In addition, it is supposed that the powder particles are randomly oriented and that the spot size on the sample is large enough so that the incident beam impinges on particles oriented along all possible crystal directions. Under these hypotheses, the SHG of the sample will be equal to that obtained from a single particle after averaging over all crystal directions. Moreover, as the light scattering is negligible, it is expected that the SHG field will be sensitive to the polarization of the incident beam. Thus, in order to analyze

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its influence on the SHG signal, we have assumed in our model that the sample is placed between two linear polarizers. For each crystal direction, all different types of wave interactions[2–4] ($oo \rightarrow o$, $oe \rightarrow o$,…) have to be considered before averaging. Here, o and e stand for ordinary and extraordinary waves, respectively.

In the case of non-phase-matchable materials, the numerical calculations show that the SHG signal oscillates as the powder particle size is varied, with a period equal to two times the average coherence length.[3] If the dispersion in the particle sizes is greater than this amount, the SHG output will be essentially independent of the powder particle size, as is often observed in practice.[20,21] As an example, Fig. 1(a) shows the theoretical results obtained for crystalline quartz
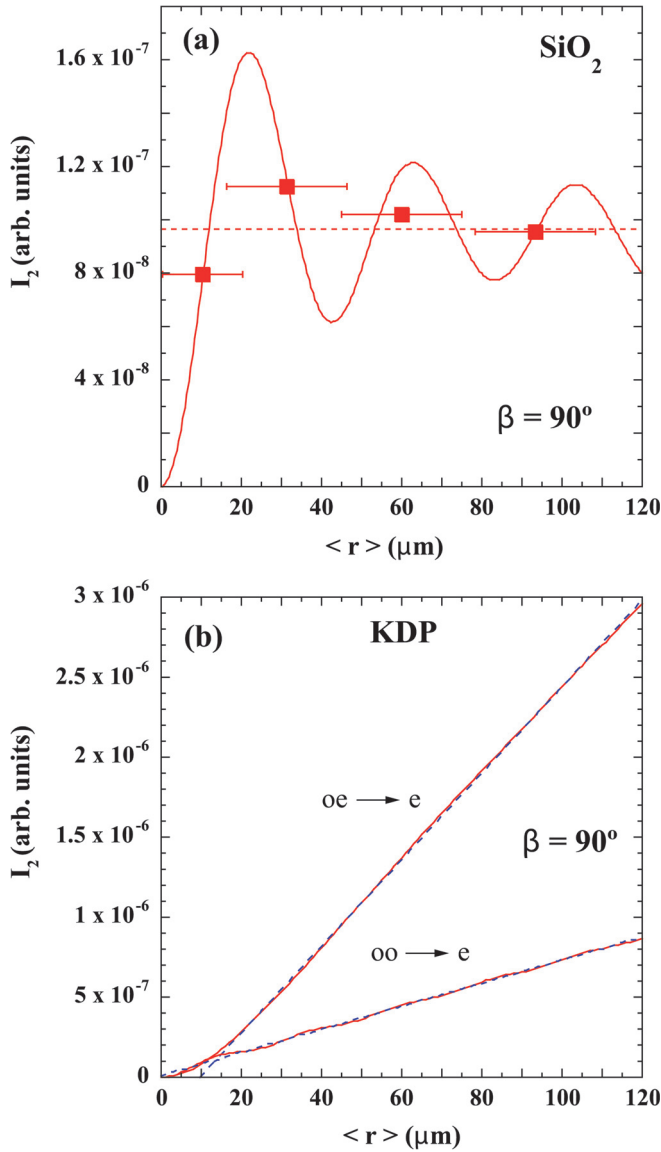

FIG. 1. (Color online) (a) Theoretical SHG intensity as a function of particle size of crystalline quartz powder for crossed polarizers (continuous line). Squares indicate the theoretical prediction assuming that the powder samples have a dispersion of particle sizes of 30 μm. The dashed line represents the SHG signal averaged over all thicknesses. (b) Theoretical SHG intensity as a function of particle size of KDP powder for crossed polarizers and $\lambda_1 = 1.064$ μm (continuous line). Both $oe \rightarrow e$ and $oo \rightarrow e$ wave interactions have been considered separately.

powder. The fundamental wavelength is 1.064 μm, and the values of the quantities required for the simulation ($n_{o,e}(\lambda_1, \lambda_2)$) were taken from Ref. 23. It can be seen that the signal oscillates with a period of about 40 μm (approximately twice the average of the coherence length[3]). However, as the particle size scatters in practice, the oscillation amplitude is greatly diminished (squares). If the dispersion is larger than 40 μm, the SHG intensity will be roughly independent of r. From that average intensity, the value of an effective nonlinear coefficient of the material can be obtained. The analysis of the SHG intensity is, however, complicated because all different types of wave interactions ($oo \rightarrow o$, $oe \rightarrow o$,…) have to be considered and, usually, all nonzero d_{ij} coefficients contribute to the SHG signal. Therefore, the data process has to be carried out numerically.

In contrast, the analysis of the SHG intensity generated by a phase-matchable material is much simpler. If the particle size is greater than about 10 μm, the light intensity generated by the wave interaction for which the PM occurs prevails over the rest. In this case, a simple analytical expression for the SHG signal can be obtained. Let us analyze, for example, the case of a negative uniaxial material ($n_o > n_e$) with a PM of type I ($oo \rightarrow e$). The SHG intensity produced by an incident beam of intensity I_1 and a wavelength λ_1 propagating through a powder particle along a crystal direction characterized by the polar and azimuthal angles (θ, φ) is

$$I_2(r, \theta, \varphi, \alpha, \beta) = b g(\alpha, \beta) \frac{d^2_{eoo}(\theta, \varphi)}{n_{2e}(\theta)} f(r, \theta), \qquad (1)$$

where

$$b \equiv \frac{8\pi^2}{\varepsilon_o c \lambda_1^2} \frac{I_1^2}{n_{1o}^2}, \qquad (2)$$

$$g(\alpha, \beta) \equiv \cos^4 \alpha \, \sin^2(\alpha + \beta), \qquad (3)$$

$$f(r, \theta) \equiv r^2 \text{sinc}^2 \left(\frac{\Delta k_{eoo}(\theta) r}{2} \right), \qquad (4)$$

$$\Delta k_{eoo}(\theta) \equiv k_e^{2\omega}(\theta) - 2k_o^\omega. \qquad (5)$$

Here, α represents the angle between the axis of the first polarizer and the eigen-axis of the fundamental ordinary wave, and β is the angle between the axes of the linear polarizers. $\Delta k_{eoo}(\theta)$ and $d_{eoo}(\theta, \varphi)$ are, respectively, the wave-number mismatch and the effective nonlinear coefficient for the interaction ($oo \rightarrow e$) considered. $n_{1o,e}$ and $n_{2o,e}$ represent the material refractive indices for the fundamental and the second harmonic waves, respectively, and r is the particle size. The SHG intensity produced by the sample can be obtained by averaging Eq. (1) over all crystal directions (θ, φ) and all relative orientations (α) of the polarization plane of the incident wave. The average over θ is mainly determined by the function $f(r, \theta)$. This function has a large peak centered at the PM angle θ_I ($\Delta k_{eoo}(\theta_I) = 0$) and successive secondary maxima with decreasing values. The peak value of the central maximum is r^2, and its angular halfwidth $\Delta \theta$ is given by $\Delta k_{eoo}(\theta_I + \Delta \theta) = 2\pi r^{-1}$. Thus, if the particle size r increases, the central peak grows sharply and narrows.

Therefore, for a large enough particle size (typically $r \geq 10~\mu m$), $f(r, \theta)$ will have significant values only for θ close to θ_I. Within that narrow angular interval, the values of the other functions of θ that appear in Eq. (1) do not appreciably change, so that in the process of averaging over θ they can be substituted by their values at θ_I. In addition, in that angular interval, the function $\Delta k_{eoo}(\theta)$ can be approximated by

$$\Delta k_{eoo}(\theta_I + \xi) \approx \left. \frac{d\Delta k_{eoo}}{d\theta} \right|_{\theta_I} \xi \equiv 2\gamma\xi, \quad (6)$$

so that

$$\int_0^{\pi/2} f(r, \theta)d\theta \approx \int_{-\infty}^{\infty} r^2 \text{sinc}^2(\gamma r\xi)d\xi = r^2 \frac{\pi}{|\gamma|r} = \frac{\pi}{|\gamma|}r. \quad (7)$$

The SHG intensity produced by the sample will be therefore given by

$$I_2(r, \beta) = \langle I_2(r, \theta, \varphi, \alpha, \beta) \rangle_{\varphi, \theta, \alpha}$$
$$= b\langle g(\alpha, \beta) \rangle_\alpha \frac{\langle d_{eoo}^2(\theta_I, \varphi) \rangle_\varphi}{n_{2e}(\theta_I)} \sin\theta_I \frac{\pi}{|\gamma|}r$$
$$= \frac{\pi^2}{4\varepsilon_o c\lambda_1} I_1^2 (3 - 2\cos 2\beta)$$
$$\times \frac{1}{\cos\theta_I n_{1o}^6 \left[(1/n_{2e}^2) - (1/n_{2o}^2) \right]} d_{eff}^2(\theta_I)r, \quad (8)$$

where

$$d_{eff}^2(\theta_I) \equiv \langle d_{eoo}^2(\theta_I, \varphi) \rangle_\varphi = \frac{1}{2\pi}\int_0^{2\pi} d_{eoo}^2(\theta_I, \varphi)d\varphi. \quad (9)$$

Proceeding in the same way, one can obtain simple analytical formulas similar to Eq. (8) for all kinds of phase-matchable uniaxial materials. In the case of a negative uniaxial material ($n_o > n_e$) with a PM of type II ($oe \rightarrow e$), for example, the SHG intensity produced by the sample will be given by

$$I_2 = \frac{2\pi^2}{\varepsilon_o c\lambda_1} I_1^2 \frac{1}{\cos\theta_{II} n_{2e}(\theta_{II}) n_{1e}(\theta_{II}) n_{1o} \left[2n_{2e}^3(\theta_{II})\left(1/n_{2e}^2 - 1/n_{2o}^2\right) - n_{1e}^3(\theta_{II})\left(1/n_{1e}^2 - 1/n_{1o}^2\right) \right]} d_{eff}^2(\theta_{II})r, \quad (10)$$

where θ_{II} represents the PM angle. It should be remarked that $d_{eff}^2(\theta_{PM})$ depends not only on the point group symmetry of the material but also on the type of PM. For example, in the case of the compound KH_2PO_4 (KDP) (negative uniaxial, point group $\bar{4}2m$), for type I PM,

$$d_{eff}^2(\theta_I) = \frac{d_{14}^2}{2}\sin^2\theta_I, \quad (11)$$

and for type II,

$$d_{eff}^2(\theta_{II}) = \frac{d_{14}^2}{2}\sin^2 2\theta_{II}. \quad (12)$$

It can be observed that in all phase-matchable uniaxial materials, the SHG output depends linearly on the powder particle size r. At first glance this result might seem surprising, because it is well known that the SHG signal produced along the PM direction grows as r^2.[2–4] Nevertheless, as we have seen, the angular width around the PM direction for which the SH is significantly emitted varies as r^{-1}, so that the SHG intensity obtained after averaging over all crystal directions depends linearly on r (see Eq. (7)). This result has been verified by numerical calculations (see Fig. 1(b)). The second harmonic intensity generated along directions (θ, φ) different from that of the PM (θ_{PM}, φ) will result in a non-null intercept of the $I_2(r)$ curve not considered in the analytical derivation carried out above. Numerical calculations confirm, however, that the slope of the real $I_2(r)$ curve is correctly given by Eqs. (8) and (10). In the case of biaxial materials, the SHG signal also depends linearly on r, but now analytical expressions like Eqs. (8) and (10) cannot be generally derived, and the analysis must be done numeri-

cally. The linear dependence I_2 versus r found in phase-matchable materials is particularly useful in practice, because the slope of the SHG curve can then be used for the experimental determination of $d_{eff}^2(\theta_{PM})$ with high accuracy (d_{14} for the $\bar{4}2m$ point group; see Eqs. (11) and (12)). In addition, the straight lines defined by Eq. (8) or (10) never become blurred by the dispersion of the particle sizes, provided that we plot I_2 versus the average particle size $\langle r \rangle$. The method is therefore insensitive to the degree of dispersion of the particle sizes in the sample.

III. EXPERIMENTAL RESULTS

In order to check the validity of these results, we carried out SHG experiments in powder crystal monolayers. We will present one example for a non-phase-matchable material (quartz) and another one for a phase-matchable material (KDP). The optical setup is described in detail in Ref. 24. A Nd:YAG laser was the fundamental light source, and the intensities generated by the different powder samples were normalized to the maximum of the first Maker fringe generated by a quartz y-cut single crystal for an $oo \rightarrow o$ conversion. The latter intensity is given by

$$I_2(SiO_2) = \frac{2}{\varepsilon_o c} I_1^2 \frac{1}{n_{2o} n_{1o}^2 (n_{2o} - n_{1o})^2} d_{11}^2(SiO_2). \quad (13)$$

Using this normalization procedure, one gets experimental access to the absolute value of the SHG signals, and, thus, d_{eff} values can be readily obtained from the normalized SHG curves.

Powders were graded using standard sieves to the desired range of particle sizes. The powder particles were

immersed in an index matching liquid and placed between two glass plates. It is important to have a reliable method to measure the grain size of the crystalline powder. Here, it is worth mentioning that, contrary to the usual belief, one cannot take r in Eq. (8) or (10) as the average value of the (consecutive) sieve sizes that permit the separation of a particular range of crystal sizes. If we model these particles in the shape of small parallelepipeds of sides $a < b < c$, then it is easy to see that the referred average value corresponds approximately to the intermediate dimension b (see Fig. 2(a)). In contrast, in the case of a monolayer, we would rather have $<r> \sim a$, simply because the parallelepipeds tend to lay on the face with the largest surface, at least in most

cases. It will be shown that in practice there can exist a big difference between $<r>$ and the value deduced from the sieve spacings. A misidentification of this parameter can give rise to an important error in the determination of the nonlinear optical coefficients.

We carried out the measurement of $<r>$ in the following way. First, we selected particles from two particular sieves. Supposedly, b will be similar to the average value of the sieve sizes, but this is irrelevant for our purposes. The $<r>$ value was deduced from the measurement of the area occupied by the crystal particles (obtained from a photograph using Adobe Photoshop software) and the volume of the particles deposited on a specific substrate area (determined from the weight of the sample and the density of the material; see Fig. 2(b) for the meaning of $<r>$ in an actual sample). The powder was deposited on a glass substrate with a circular mask of an area of about 0.5 cm². A vibrator was used to spread the powder uniformly. Figure 2(c) shows a photograph of a KDP sample with particles that were selected to be smaller than 62 μm. The value deduced for $<r>$ was 9 μm, which is rather far from the intermediate dimension of 31 μm. Figure 2(d) shows some $<r>$ values of different KDP powders as a function of b. It can be seen that in all cases, $<r>$ is smaller than b. Also, it is interesting to point out that in no case do we make any assumption about the r distribution or about the shape of the particles, but we obtain $<r>$ operationally each time. In addition, it can be understood that the measurement of $<r>$ is rather accurate, with the accuracy being limited mainly by the measurement of the percentage of occupied area on the photograph (error typically $< 5\%$). Finally, to normalize the SHG signal with that of the reference single crystal, the intensity of each powder sample must be linearly extrapolated to that of a sample with a 100% occupation fraction.
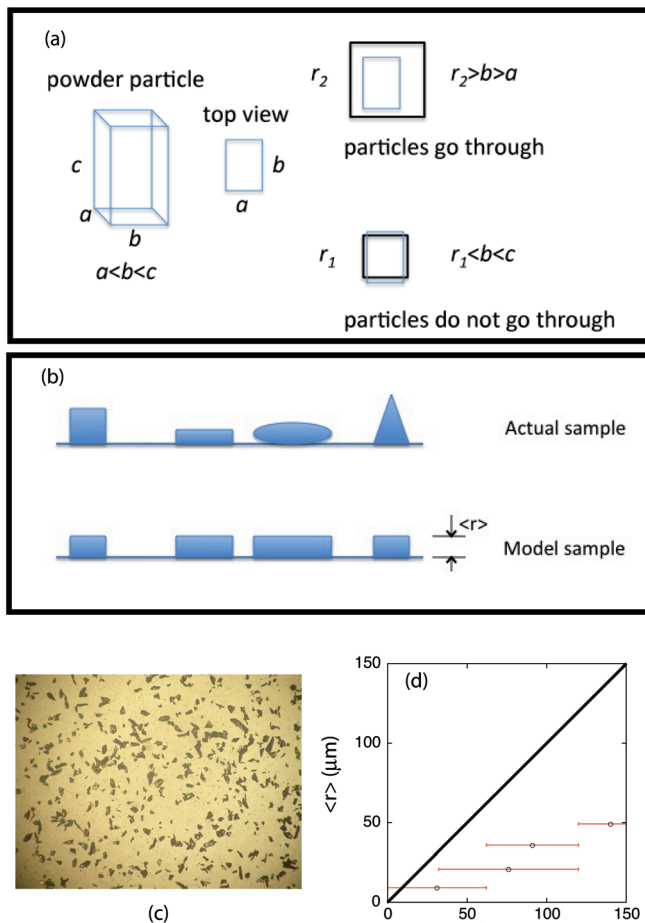

FIG. 2. (Color online) (a) Schematic diagram of the sieve process for crystal particles. For simplicity, the particles are assumed to be orthogonal parallelepipeds of dimensions $a < b < c$. If the particle is selected between two sieves of mesh spacings r_2 and r_1 ($r_2 > r_1$), this means that $r_2 > b > a$ and $r_1 < b < c$. However, it is not known whether $r_2 < c$ or $r_2 > c$, or whether $r_1 > a$ or $r_1 < a$. Thus we can deduce that, approximately, $b \sim (r_2 + r_1)/2$, i.e., the intermediate dimension is the one that can be selected in advance during the sieve operation. (b) Schematic view of the powder particles in an actual sample (arbitrary grain shapes are possible). $<r>$ is the thickness of the parallelepipeds of the simplified sample that correspond to the correct volume of the actual sample. In both cases, the area occupied by the grains is the same. (c) Photomicrograph of a KDP powder sample obtained by using a single sieve with a spacing of 62 μm. The area occupied by the crystal is 18%, and the average thickness of the sample is $<r> = 9$ μm. In order to compare the SHG signal with that of the single crystal of quartz, the intensity must be extrapolated to 100%. Image width: 1.4 mm. (d) Particle size $<r>$ as a function of the average of the consecutive sieve spacings for KDP powders.
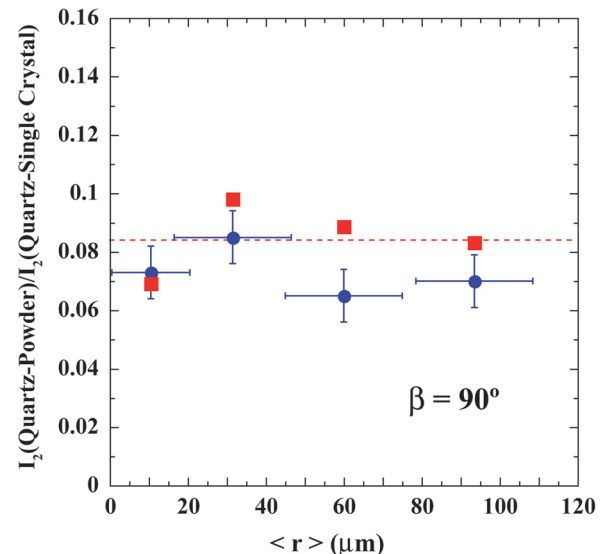

FIG. 3. (Color online) SHG intensity measured in crystalline quartz powder as a function of particle size for crossed polarizers (dots), and the theoretical prediction (squares) assuming $d_{11} = 0.30$ pm/V and a dispersion of particle sizes of 30 μm. The dashed line represents the theoretical SHG signal averaged over all thicknesses. The vertical scale is normalized with respect to the maximum of the SHG intensity (Maker fringe) of a quartz single crystal that undergoes an $oo \rightarrow o$ SHG conversion.

The dots in Fig. 3 represent the experimental data for quartz powder, and the squares represent the theoretical prediction, assuming $d_{11} = 0.30$ pm/V and a dispersion of particle sizes of 30 μm. All intensities were normalized to the SHG value of a single crystal of quartz using Eq. (13), as explained above. As can be seen, the data are almost independent of $<r>$, as should correspond to a material showing no PM if the width of the particle-size distribution is comparable to twice the coherence length (in our case, we estimate a distribution width of at least 30 μm, from the difference of the consecutive sieve sizes). From the average value of the experimental data, we obtained $d_{11} = 0.28 \pm 0.02$ pm/V for the quartz powder, assuming a reference[25] $d_{11}(SiO_2) = 0.30$ pm/V.

The results obtained with the KDP powders are shown in Fig. 4. At the fundamental wavelength used in the experiment, both types of PM coexist,[6,25] so the total SHG intensity will be described by the sum of Eqs. (8) and (10). Because the term of type I depends on the angle between polarizers β and the term of type II does not, it is possible to separate out the contribution of each term to the total intensity by changing the angle β. In Figs. 4(a) and 4(b), the experimental data obtained with parallel ($\beta = 0°$) and crossed ($\beta = 90°$) polarizers, respectively, are displayed. From the values of the refractive indices for $\lambda_{1,2}$ and the slope of the line that best fits the experimental points, the value of the $d_{14}(KDP)$ coefficient can be determined using Eqs. (8) and (10). Thus, assuming[25] $d_{11}(SiO_2) = 0.30$ pm/V, we obtain $d_{14}(KDP) = 0.43 \pm 0.03$ pm/V from the data of Fig. 4(a), and $d_{14}(KDP) = 0.40 \pm 0.02$ pm/V from those of Fig. 4(b). Both values are in excellent agreement with that reported in the literature.[6,25,26] The model developed can correctly describe not only the slope of the fitting lines but also the value of the SHG intensity itself (squares in Fig. 4). However, for this purpose the SHG intensity must be obtained numerically.

IV. CONCLUSION

In summary, an accurate procedure for determining the d_{ij} coefficients from powder crystal monolayers is proposed. The method overcomes some major limitations of the conventional powder method proposed by Kurtz and Perry,[7] particularly those related to the light scattering inherent in a thick sample. It has been demonstrated that this method can provide reliable and accurate values of the coefficients of both phase-matchable and non-phase-matchable materials. In the case of phase-matchable uniaxial materials, the analysis is particularly straightforward, because the SHG intensity is well described by simple analytical formulas. In addition, in this case, the d_{eff} measurement is not affected by the unavoidable dispersion of the particle size in actual samples. Therefore, the method constitutes a real alternative to the single crystal techniques in those cases where it is difficult to grow large crystals.
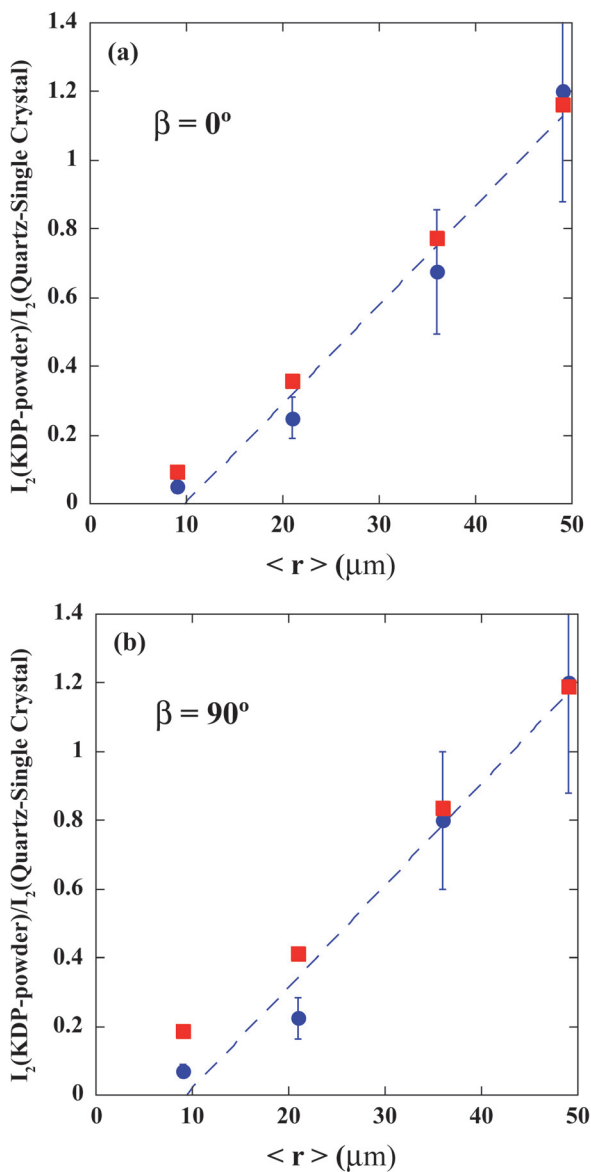
FIG. 4. (Color online) SHG intensity from a monolayer of KDP powder as a function of the particle size for (a) parallel polarizers ($\beta = 0°$) and (b) crossed polarizers ($\beta = 90°$). The intensities have been normalized with respect to the maximum intensity generated by a quartz single crystal for an $oo \to o$ interaction. Dots represent the experimental data, and squares represent those numerically obtained from the model. The dashed line is a linear least squares fit to the experimental points.

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