

Application of polycrystalline diamonds for magnetometry based on interactions of nonequivalently oriented groups of NV centres

R.A. Akhmedzhanov, L.A. Gushchin, I.V. Zelensky, V.A. Nizov, N.A. Nizov, D.A. Sobgaida

Abstract. The possibility of using polycrystalline diamonds for measuring weak magnetic fields based on cross-relaxation interaction between nonequivalently oriented NV-centre groups is theoretically shown and experimentally demonstrated.

Keywords: NV centre, magnetometry, cross relaxation, polycrystalline diamond.

The unique properties of negatively charged nitrogen-vacancy (NV) centres in diamond make them attractive for various applications, among which magnetometry is most promising and ready for commercial use [1]. A standard magnetometry scheme using NV centres is based on optically detected magnetic resonance (ODMR) [1–4]. This method consists in observation of dips appearing in the optically (usually nonresonantly) excited fluorescence of NV centres upon scanning the microwave radiation frequency and can be used in a wide temperature range, from cryogenic temperature [5, 6] to 700 K [7]. The possibility of operation with both single defects and their ensembles makes it possible to select an optimal combination of spatial resolution and measurement accuracy for a particular problem. A substantial limitation of this approach is the use of microwave radiation, which restricts the spectrum of possible applications.

Recently, some magnetometry schemes with NV centres without using microwave radiation have been proposed. To measure strong fields, it was suggested to use the effect of a decrease in the fluorescence intensity [8] and a change in the spin relaxation contrast [9] in a magnetic field, but these methods have a relatively low sensitivity. A protocol for measuring weak magnetic fields based on anticrossing of spin sublevels of the ground state of NV centres was proposed in [10]. This protocol requires application of an additional strong (~ 1000 G) magnetic field, which somewhat restricts the range of its application. A promising but rather complicated method is based on the effect of electromagnetically induced transparency [11].

R.A. Akhmedzhanov, L.A. Gushchin, I.V. Zelensky, V.A. Nizov, N.A. Nizov, D.A. Sobgaida Institute of Applied Physics, Russian Academy of Sciences, ul. Ulyanova 46, 603950 Nizhny Novgorod, Russia; e-mail: zelensky@appl.sci-nnov.ru

Received 11 June 2018; revision received 23 July 2018
Kvantovaya Elektronika 48 (10) 912–915 (2018)
Translated by M. N. Basieva

In [12], we proposed and experimentally demonstrated a method for magnetic field measurement based on the observation of cross-relaxation resonances in NV centres in diamond single crystals. The method consists in the following. The axis of NV centres in diamond crystal can be oriented along four different directions, namely, $[1, 1, 1]$, $[-1, 1, 1]$, $[1, -1, 1]$, and $[1, 1, -1]$. The frequencies of transitions between the ground state sublevels depend on the magnetic field strength and direction and, in general, do not coincide for NV centres with different orientations. At some magnetic field directions, the transition frequencies for one or several groups of NV centres may coincide, which will lead to cross-relaxation interaction between them and to a change in the signal of optically induced fluorescence.

Let a magnetic field be a sum of an unknown constant field and a scanned field. In this case, the dependence of the fluorescence intensity on the scanned field may exhibit up to nine cross-relaxation resonances. The resonance positions depend on the constant magnetic field and, therefore, can be used for its measurement. The proposed method ensures a high measurement accuracy and does not require microwave radiation, strong magnetic fields, etc. However, for precision measurements, it is necessary to highly accurately orient the scanned magnetic field with respect to the crystal axes. In addition, some problems may arise with measuring weak magnetic fields insufficient for complete splitting of resonances.

In the present work, we consider the possibility of using polycrystalline diamonds for magnetometry based on cross-relaxation interaction of NV centres. Note that the possibility of using polycrystalline samples and nanocrystals with random axis orientations for magnetometry was also discussed in works [13, 14].

In the case of a polycrystalline sample with random orientations of axes, there is no any preferable direction and, hence, the fluorescence signal can depend only on the magnetic field strength: $F(\mathbf{B}) = F(B)$. Dependence $F(B)$ was calculated using numerical simulation. The energy of dipole–dipole interaction between NV centres in diamond rapidly decreases with increasing distance between them, because of which the simulation was performed assuming that NV centres with different axis orientations interact in pairs and all the possible (six) pairs contribute with identical weights to the total fluorescence signal. When modelling the fluorescence from a pair of interacting NV centres, we assumed that cross relaxation leads to their depolarization, because of which the fluorescence signal under the conditions of nonresonance pumping decreases [1, 12]. It was supposed that the intensity of cross-

relaxation processes depends only on the difference between the frequencies of transitions between sublevels of the ground state of NV centres. The ground state of the NV centres is a triplet, its sublevels with $m = 0$ and ± 1 being split by ~ 2.88 GHz at a zero magnetic field. For simplicity, we summed the contributions from all the possible interactions at the $|0\rangle \leftrightarrow |+1\rangle$ and $|0\rangle \leftrightarrow |-1\rangle$ transitions. Possible interactions via the $|0\rangle \leftrightarrow |+1\rangle$ and $|-1\rangle \leftrightarrow |+1\rangle$ transitions were not taken into account because their frequencies can coincide only in relatively strong fields. The interaction via the $|-1\rangle \leftrightarrow |+1\rangle$ transitions of different centres was also ignored because these transitions are forbidden in the dipole approximation. The dependence of the fluorescence signal on the difference between transition frequencies $\Delta\omega$ was described by the Lorentzian profile

$$F(\Delta\omega) = 1 - C \left[1 + \left(\frac{\Delta\omega}{\omega_c} \right)^2 \right]^{-1}. \quad (1)$$

Contrast C and characteristic width ω_c were chosen so that the calculation results for a single crystal sample corresponded to the experimental results obtained in [12]. For frequencies ω_+ and ω_- of transitions $|0\rangle \leftrightarrow |+1\rangle$ and $|0\rangle \leftrightarrow |-1\rangle$, we used the expression [12, 15]

$$\omega_{\pm} = \frac{2\sqrt{3}D}{3}(3b^2 + 1)^{1/2} \sin\left(\frac{\alpha}{3} + \delta_{\pm}\right), \quad (2)$$

where $\delta_+ = \pi/3$, $\delta_- = 2\pi/3$; $b = \gamma\mu_B B/D$ is the dimensionless magnetic field, $D \approx 2.88$ GHz, and parameter α is determined by the expression

$$\cos\alpha = \frac{9b^2 - 27b^2 \cos^2\theta + 2}{2(3b^2 + 1)^{3/2}} \quad (3)$$

(θ is the angle between the magnetic field and the axis of the NV centre).

The dependence of the fluorescence signal intensity on the magnetic field strength was calculated for a particular magnetic field orientation with respect to the crystallographic axes of the sample. Then, we performed averaging over a large number of random orientations.

The calculation results are presented in Fig. 1a. The fluorescence signal dependence for a polycrystalline sample is shown by the solid curve. For comparison, the dashed and dotted lines show the dependences calculated for a monocrystalline sample and different magnetic field directions (along vectors $[1, 2, 3]$ and $[1, 0.25, 0.1]$, respectively). One can see that the use of a polycrystalline sample causes no noticeable increase in the width of the cross-relaxation resonance. Moreover, resonance in monocrystalline diamond for some magnetic field directions can be even wider. It is necessary to note that the resonance in the case of polycrystals has much ‘heavier’ wings, which, nevertheless, does not affect the accuracy of determination of the resonance peak position.

Following [12], let us represent the magnetic field in the form of a sum of the measured (\mathbf{B}_0) and scanned (\mathbf{B}_{sc}) magnetic fields. In this case, the dependence of the fluorescence signal on the scanned magnetic field strength will be written as

$$f(B_{sc}) = F\left(\sqrt{(B_{sc} + B_{\parallel})^2 + B_{\perp}^2}\right), \quad (4)$$

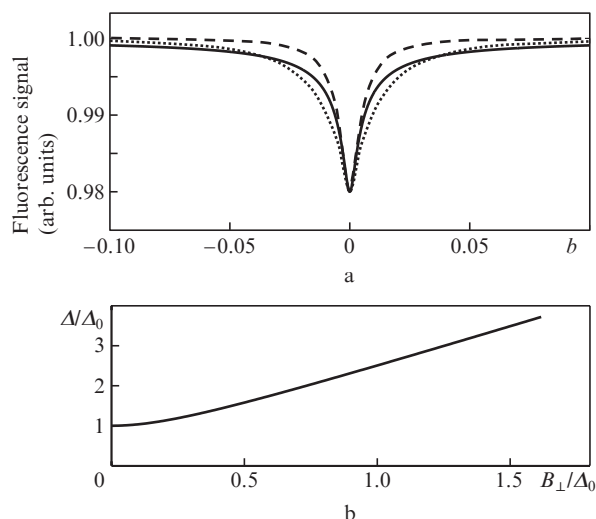


Figure 1. (a) Dependences of the fluorescence signal on dimensionless magnetic field for (solid curve) polycrystalline and (dashed and dotted curves, which correspond to different magnetic field directions) single crystal samples and (b) dependence of the resonance HWHM Δ on the transverse magnetic field B_{\perp} . Δ_0 is the cross-relaxation resonance half-width in the absence of a transverse magnetic field.

where B_{\parallel} and B_{\perp} are the longitudinal and transverse (with respect to the scanned magnetic field) components of the measured field.

The fluorescence signal profile will exhibit a single cross-relaxation resonance with a centre at

$$B_{sc} = -B_{\parallel}. \quad (5)$$

Thus, the measurement procedure is reduced to recording the cross-relaxation resonance position, which allows one to directly determine the measured field projection on the scanned field direction. If necessary, one can perform several measurements with different directions of \mathbf{B}_{sc} and determine the magnetic field strength and direction.

The transverse magnetic field component does not affect the resonance position but leads to its broadening and a decrease in contrast. From (4), it follows that the fluorescence signal in the cross-relaxation resonance centre depends only on B_{\perp} : $F(B_{\perp})$. We will assume that $F(B = \infty) = 1$; then, the expression for contrast can be written as

$$C(B_{\perp}) = 1 - F(B_{\perp}). \quad (6)$$

From this it follows that the resonance contrast decreases twofold at $B_{\perp} = \Delta_0$, where Δ_0 is the cross-relaxation resonance half-width in the absence of a transverse magnetic field. The broadening of the cross-relaxation resonance caused by a transverse field can also be calculated based on formula (4). The dependence of resonance HWHM Δ on the transverse magnetic field is presented in Fig. 1b. One can see that the resonance for weak fields ($B_{\perp} \ll \Delta_0$) almost does not broaden, while Δ at strong transverse fields is proportional to B_{\perp} .

Thus, a decrease in contrast and the broadening of the cross-relaxation resonance caused by a transverse field can be considered acceptable for transverse fields with strengths below or about the unperturbed resonance half-width (in our experiments, this value is ~ 5 G).

To check the theoretical concepts developed in the present study, we performed an experiment using the scheme presented in Fig. 2a. As a polycrystalline sample, we used a stack of ~ 25 glued diamond crystals (HPHT diamonds with a characteristic size of ~ 0.25 mm irradiated by an electron beam with an intensity of 10^{18} electron cm^{-2} and then annealed at a temperature of 800°C) on a glass substrate. The transverse size of the stack was ~ 2 mm. Excitation was performed by a laser with a wavelength of 532 nm, a beam width exceeding the transverse size of the batch, and a radiation intensity of 8 W cm^{-2} . The fluorescence was collimated by a lens and detected by a photodiode; the residual pump radiation was cut off by a light filter. Due to a small number of crystals in the stack, for better imitation of a polycrystal, we performed measurements at several (eleven) rotations of the sample around the longitudinal axis and averaged the measurement results. The sample was placed in a scanned magnetic field formed by a solenoid. An additional magnetic field directed at an angle to the scanned field was applied to the sample using a magnetic coil (not shown in Fig. 2a).

The dependences of the fluorescence intensity on the scanned magnetic field for different currents through the magnetic coil are shown in Fig. 2b. The longitudinal component of the additional magnetic field causes a shift of the cross-relaxation resonance, while the transverse component leads to a decrease in the contrast and to a broadening. The irregular shape of the resonance is related to a small number of crystals in the stack and, according to calculations, can be considerably improved by increasing

the number of crystals in the working volume above 1000, which can be easily achieved by using polycrystalline diamond samples.

Figure 2c shows the dependence of the projection of the additional magnetic field on the coil current, which was measured by the cross-relaxation resonance position. The measured results coincide to a high accuracy with a linear dependence. Upon measurements, the transverse component of the additional magnetic field invariably exceeded the longitudinal component (the angle between the scanned and additional fields was larger than 45°). Our experiments confirm the feasibility of the proposed method for measuring magnetic fields up to several gauss.

Thus, in the present work, we theoretically showed and experimentally demonstrated the possibility of using polycrystalline diamonds for measuring magnetic fields based on the cross-relaxation between nonequivalently oriented NV-centre groups. The proposed method can be used for magnetic fields up to several gauss. The use of polycrystalline samples instead of diamond single crystals allows simplification of the measurement procedure. In particular, in the case of polycrystalline samples, precise orientation of the axes is not needed and one observes only one cross-relaxation resonance whose position makes it possible to directly determine the magnetic field projection on a given direction. Note also that the use of polycrystalline diamonds and diamond powders instead of single crystals may considerably reduce the magnetometer cost.

Acknowledgements. We are grateful to V.L. Velichansky and A. O. Levchenko for providing the diamond samples. The theoretical part of the study was supported by the Russian Foundation for Basic Research and the Government of the Nizhny Novgorod region (Project No. 18-42-520017); the experimental part of the work was supported by the State Assignment for Scientific Research (Theme No. 0035-2014-0005).

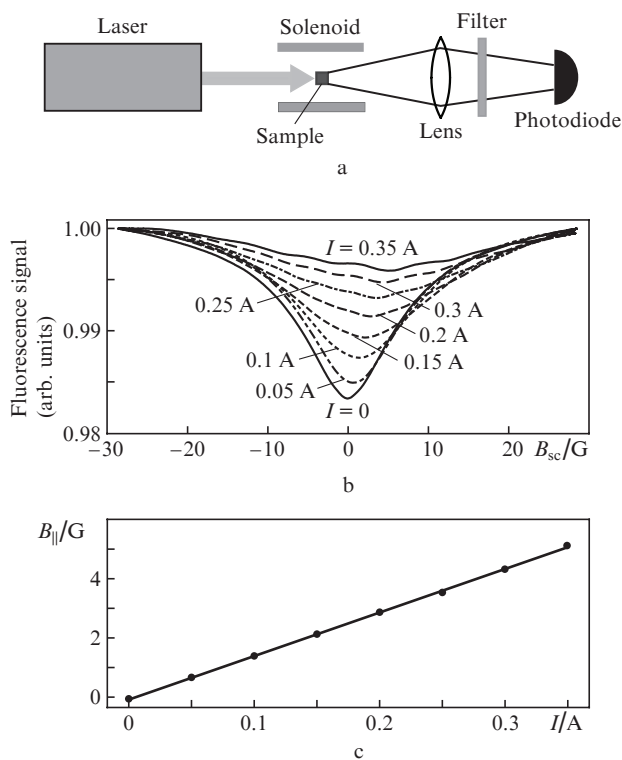


Figure 2. (a) Experimental scheme, (b) cross-relaxation resonance at different magnetic coil currents I , and (c) dependence of the longitudinal magnetic field component measured by the cross-relaxation resonance position on the magnetic coil current and its linear approximation.

References

1. Rondin L., Tetienne J.-P., Hingant T., Roch J.-F., Maletinsky P., Jacques V. *Rep. Progr. Phys.*, **77**, 056503 (2014).
2. Taylor J.M., Cappellaro P., Childress L., Jiang L., Budker D., Hemmer P.R., Yacoby A., Walsworth R., Lukin M.D. *Nature Phys.*, **4**, 810 (2008).
3. Maze J.R., Stanwix P.L., Hodges J.S., Hong S., Taylor J.M., Cappellaro P., Jiang L., Gurudev Dutt M.V., Togan E., Zibrov A.S., Yacoby A., Walsworth R.L., Lukin M.D. *Nature*, **455**, 644 (2008).
4. Pham L.M., Le Sage D., Stanwix P.L., Yeung T.K., Glenn D., Trifonov A., Cappellaro P., Hemmer P.R., Lukin M.D., Park H., Yacoby A., Walsworth R.L. *New J. Phys.*, **13**, 045021 (2011).
5. Thiel L., Rohner D., Ganzhorn M., Appel P., Neu E., Muller B., Kleiner R., Koelle D., Maletinsky P. *Nature Nanotechnol.*, **11**, 677 (2016).
6. Pelliccione M., Jenkins A., Ovartchaiyapong P., Reetz C., Emmanouilidou E., Ni N., Bleszynski Jayich A.C. *Nature Nanotechnol.*, **11**, 700 (2016).
7. Toyli D.M., Christle D.J., Alkauskas A., Buckley B.B., Van de Walle C.G., Awschalom D.D. *Phys. Rev. X*, **2**, 031001 (2012).
8. Rondin L., Tetienne J.-P., Spinicelli P., Dal Savio C., Karrai K., Dantelle G., Thiaville A., Rohart S., Roch J.-F., Jacques V. *Appl. Phys. Lett.*, **100**, 153118 (2012).
9. Simpson D.A., Tetienne J.-P., McCoe J.M., Ganesan K., Hall L.T., Petrou S., Scholten R.E., Hollenberg L.C.L. *Sci. Rep.*, **6**, 22797 (2016).

10. Wickenbrock A., Zheng H., Bougas L., Leefers N., Afach S., Jarmola A., Acosta V.M., Budker D. *Appl. Phys. Lett.*, **109**, 053505 (2016).
11. Acosta V.M., Jensen K., Santori C., Budker D., Beausoleil R.G. *Phys. Rev. Lett.*, **110**, 213605 (2013).
12. Akhmedzhanov R., Gushchin L., Nizov N., Nizov V., Sobgayda D., Zelensky I., Hemmer P. *Phys. Rev. A*, **96**, 013806 (2017).
13. Anisimov A.N., Babunts R.A., Kidalov S.V., Mokhov E.N., Soltamov V.A., Baranov P.G. *JETP Lett.*, **104**, 82 (2016).
14. Fedotov I.V., Doronina-Amitonova L.V., Sidorov-Biryukov D.A., Safronov N.A., Levchenko A.O., Zibrov S.A., Blakley S., Perez H., Akimov A.V., Fedotov A.B., Hemmer, P., Sakoda K., Velichansky V.L., Scully M.O., Zheltikov A.M. *Opt. Lett.*, **39**, 6755 (2014).
15. Holliday K., Manson N.B., Glasbeek M., van Oort E. *J. Phys.: Condens. Matter*, **1**, 7093 (1989).